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No. NW-T-1.7

Waste from Innovative Types of Reactors and Fuel Cycles

A Preliminary Study

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WASTE FROM INNOVATIVE TYPES OF REACTORS AND FUEL CYCLES

A PRELIMINARY STUDY

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FOREWORD

One of the IAEA's statutory objectives is to "seek to accelerate and enlarge the contribution of atomic energy to peace, health and prosperity throughout the world." One way this objective is achieved is through the publication of a range of technical series. Two of these are the IAEA Nuclear Energy Series and the IAEA Safety Standards Series.

According to Article III.A.6 of the IAEA Statute, the safety standards establish "standards of safety for protection of health and minimization of danger to life and property". The safety standards include the Safety Fundamentals, Safety Requirements and Safety Guides. These standards are written primarily in a regulatory style, and are binding on the IAEA for its own programmes. The principal users are the regulatory bodies in Member States and other national authorities.

The IAEA Nuclear Energy Series comprises reports designed to encourage and assist R&D on, and application of, nuclear energy for peaceful uses. This includes practical examples to be used by owners and operators of utilities in Member States, implementing organizations, academia, and government officials, among others. This information is presented in guides, reports on technology status and advances, and best practices for peaceful uses of nuclear energy based on inputs from international experts. The IAEA Nuclear Energy Series complements the IAEA Safety Standards Series.

In generic terms, sustainable development implies not just efficient production practices and efficient use of natural resources but also a reduction of toxic solid, liquid and gaseous emissions. The nuclear industry is planning for the introduction of innovative nuclear energy systems with enhanced safety profiles. These new reactor systems and their associated open and closed fuel cycles hold significant potential for enhanced sustainability through a further reduction of CO_2 emissions and enhanced efficiencies in energy production and resource use. These new systems also offer considerable scope for the drastic minimization of waste volume and radiotoxicity that is consistent with the precepts on enhanced producer responsibility.

This preliminary publication sets the stage for considering the back end of the fuel cycle and waste generation early in the development of new reactors and their associated fuel cycles. Given that, currently, these advanced reactors exist only as concepts, and since fuel types for these reactors are not yet totally defined, it is difficult to be precise about the chemical and physical nature of the irradiated fuel or the operational wastes and wastes from fuel recycling. Nevertheless, it is possible to describe these waste flows in broad chemical and physical terms and identify possible processing, recycling and disposition pathways. This constitutes the primary objective of this publication. In taking an early and integrated approach, the nuclear industry is taking an opportunity to become a forerunner in extended producer responsibility.

The IAEA officers responsible for this publication were Z. Drace, A. Ponomarev and K. Qureshi of the Division of Nuclear Power.

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CONTENTS

1. INTRODUCTION

1.1. BACKGROUND

Today there are 454 nuclear power reactors in operation in 30 countries around the world, accounting for approximately 13% of electricity generation worldwide. Another 54 nuclear power reactors are currently under construction in 15 countries. Nuclear power has an excellent operating record and generates electricity in a reliable, environmentally safe and affordable manner. Concerns over energy resource availability, climate change, air quality and energy security suggest an important role for nuclear power in future energy supplies. While current nuclear power plant (NPP) designs provide an economically and publicly acceptable electricity supply in many markets, further advances in innovative nuclear energy system (INS) design can broaden opportunities for the use of nuclear energy.

Innovative power reactors and fuel cycles are generally not expected to be available for commercial construction before 2030. Various groups are conducting research into these reactor types, including the Generation IV International Forum (GIF) [1] and the IAEA's International Project on Innovative Nuclear Reactors and Fuel Cycles (INPRO). The primary goals of these innovative reactors and fuel cycles are to enhance nuclear safety, improve proliferation resistance, minimize waste and natural resource utilization, and decrease the cost to build and run such plants.

When considering the waste minimization aspects of INSs, it should be noted that the total amount of waste generated from the operation of a nuclear power reactor consists not only of the operational reactor waste, but of waste arising from its decommissioning, waste from the front end nuclear fuel cycle generated at all stages of fuel manufacture, and waste from the back end nuclear fuel cycle, which may include spent fuel encapsulated for direct disposal or waste arising from the reprocessing of spent fuel if the closed fuel cycle option is chosen. The amount of waste generated affects the economics of nuclear power, and the radioactive component of this waste has an impact on human health and the environment, particularly in the long term. The chemical-physical composition of the waste determines the selection of technology for its processing.

Minimization of waste is an important consideration from the economic viewpoint, while changing the radioactive and chemical–physical composition of waste may notably reduce the long term stewardship burden in the future, thereby improving protection for public health and the environment. Any radioactive waste, independent of its origin, must be managed in accordance with internationally accepted fundamental waste management principles [2]. In addition, the sustainability of the waste management system should be assessed using INPRO methodology.

The waste that is produced can be treated to reduce the volume requiring disposal. The most desirable way to minimize waste is to reduce it at the source; however, operational demands for effectiveness and efficiency limit the feasibility of this approach. As part of an integrated waste management strategy, all aspects of reactor design and operation should be reviewed to identify optimal possibilities for reducing the future impact of the waste produced (e.g. volume, radiotoxicity, thermal load). Considering each component individually rather than as an integrated system may produce suboptimal results.

The reduction of radiotoxicity is a valid metric for long term impacts on waste management; it is heavily based on actinide content. However, these actinides are not mobile in a repository environment. The substances of concern which dominate the dose calculations in any repository design are long lived, mobile fission and activation products such as ^{129}I, ^{14}C and ^{99}Tc. From a human health risk standpoint, chemical toxicity is more important than reduction of radiotoxicity. This will not decrease over time. In some cases, advanced fuel cycles end up increasing the chemical toxicity of the waste. There will always be a trade-off between the potential long term dose from a repository vs. the immediate, real dose from operations of a fuel cycle facility (e.g. emissions to the environment).

Waste management systems for innovative reactors and fuel cycles should ensure optimal protection of human health and the environment free from exposure to radiation and harmful chemicals. Factors to be considered include:

- Radiotoxicity of the waste generated, as a function of time;
- Ability of the waste form to retain radionuclides (especially mobile and long lived) under normal and accident conditions;
- Mobility of the chemically toxic elements through environmental pathways;

- Length of time that waste containing long lived radionuclides remains in interim storage;
- Degree to which the waste is kept in a passively safe state;
- Occupational exposure in waste management facilities.

For existing reactors and fuel cycles, the expected types of waste to be generated are known, and aside from a few potentially problematic wastes (e.g. graphite, tritium, ^{14}C), most have clearly defined cradle to grave (end to end) pathways. For new and innovative reactors and fuel cycles, however, some waste types will be either new or known to be problematic. A primary challenge associated with developing innovative reactors and fuel cycles is to ensure that all problematic waste streams are identified and efforts are initiated to find solutions well before the waste streams are expected to reach the end state.

Accordingly, it is necessary to identify the problematic waste streams that will be generated from nuclear fuel cycles supporting the operation of these reactors and to develop consensus on the optimal approach(es) for management of such waste.

1.2. OBJECTIVES

The objectives of this publication are to:

- Describe a systematic approach for identifying the types of waste expected to be generated from innovative reactor designs and corresponding nuclear fuel cycles during both routine and abnormal operating conditions (or during forced outage), with emphasis on potentially problematic wastes (i.e. wastes which are substantially different from those produced by the current reactors and fuel cycles);
- Provide examples of waste streams identified by this approach;
- Discuss the technology needed to process and store waste, and identify problematic waste for which processing technology has yet to be developed;
- Identify known or potential severe challenges or showstoppers related to further development of approaches to manage wastes resulting from innovative reactors and fuel cycles;
- Recommend further research and studies.

The above objectives are aimed at supporting the systematic evaluation of innovative reactors and fuel cycles (e.g. via INPRO).

1.3. SCOPE

This publication focuses on three areas:

- Identification of principal waste streams from innovative reactors and fuel cycles;
- Identification of the optimal disposition (processing and disposal) pathways for the most common of these wastes (e.g. combustible, compactable, ferrous metals, resins, sludge);
- Identification of any problematic waste from innovative reactor designs and fuel cycles which will require further study as part of a separate project.

The third component of the scope is critical, as it defines the significant long term challenges to innovative reactors and fuel cycles from the waste management perspective.

Since this publication is concerned with wastes generated by innovative nuclear power reactors and fuel cycles whose designs are continuing to evolve, a qualitative approach is adopted.

1.4. STRUCTURE

Section 2 reviews the various innovative nuclear reactors and fuel cycles (with emphasis on the reprocessing of nuclear fuel). Section 3 summarizes the goals and methodology for assessing the waste from innovative nuclear reactor systems. Section 4 identifies the principal waste streams arising from operating innovative nuclear reactors. Section 5 assesses the probable waste streams from future decommissioning of innovative nuclear reactors and fuel cycle facilities. Section 6 summarizes the waste management issues for innovative nuclear reactors and fuel cycles. Section 7 contains some conclusions and recommendations for future research and development.

The publication also contains several annexes. Annex I briefly introduces the stages of a conventional nuclear fuel cycle based on uranium and partially mixed (uranium–plutonium) oxide (MOX) fuel. Annex II describes in more detail examples of innovative nuclear power reactors. Annex III describes in more detail examples of innovative nuclear fuel cycles, while Annex IV describes the evolution of the approach to spent nuclear fuel (SNF) reprocessing in France.

2. INNOVATIVE NUCLEAR POWER REACTORS AND FUEL CYCLES

A number of projects and studies have been launched worldwide to further develop nuclear energy technology in the near future (15–25 years) and beyond. It is widely recognized that future development must consider the competing demands of sustainability, economic viability, safety, environmental protection, waste management and proliferation resistance. The INPRO programme at the IAEA develops methodologies to systematically take into account each of these factors.

A nuclear energy system comprises the complete spectrum of nuclear facilities and associated institutional measures. Nuclear facilities include infrastructure for mining and milling, processing and enrichment of uranium and/or thorium, manufacturing of nuclear fuel, production (of electricity or other energy supply), reprocessing of nuclear fuel, and facilities for management activities of related materials, including transportation and waste management. Institutional measures consist of agreements, treaties, national and international legal frameworks and conventions (such as the Treaty on the Non-Proliferation of Nuclear Weapons, the International Nuclear Safety Convention and IAEA Safeguards Agreements) and the national and international infrastructure needed to operate a nuclear programme.

In supporting future nuclear energy systems, INPRO considers all types of reactors, such as:

- Reactors cooled by light water and heavy water, gas, liquid metal and molten salt;
- Reactors of different sizes of thermal power and use (e.g. production of electricity, process and district heat and freshwater);
- Reactors for partitioning and transmutation of fission products;
- Reactors with associated fuel cycles (e.g. U, U–Pu, Th, U–Pu–Th).

All phases in the life cycle of such facilities are included: site acquisition, design, construction, equipment manufacturing and installation, commissioning, operation, decommissioning and site release/closure. An example of such a nuclear energy system could be a thermal reactor operating in an open fuel cycle, a fast reactor operating in a closed fuel cycle, or a combination of thermal reactors and fast reactors in a closed fuel cycle.

INS refers to systems that will position nuclear energy to make a major contribution to the global energy supply in the twenty-first century. In this context, future systems may include evolutionary as well as innovative designs. An evolutionary design [3] is an advanced design that achieves improvements over existing designs through small to moderate modifications, with a strong emphasis on maintaining proven design concepts to minimize technological risks. An innovative design [3] is an advanced design which incorporates radical conceptual changes in design approaches or system configuration in comparison with existing practice. These systems comprise not only electricity generating plants, but also include plants (of various sizes and capacities) for other applications — such as high temperature heat production, district heating and sea water desalination — to be deployed in developed regions as well as in developing countries and countries in transition.

Innovative reactors are being studied under a variety of international initiatives and collaborative projects — such as GIF, INPRO, the European Union's Sustainable Nuclear Energy Technology Platform (SNETP) programmes and various national activities [1, 4–16]. Figure 1 shows the basic idea of GIF.

With respect to waste management, INSs are anticipated to generate less waste than existing designs per unit of energy generated (e.g. waste/kWh(e)), with reduced radiotoxicity, fewer long lived actinides and a smaller footprint for geological disposal. This development will facilitate larger scale application of nuclear energy with less impact to the public and the environment.

2.1. INNOVATIVE NUCLEAR POWER REACTORS

A variety of thermal and fast reactor systems have been identified in various programmes as potential candidates for future deployment. The basic technologies and applicable fuel cycles are summarized in Table 1.

All fast reactors feature a fast neutron spectrum and operate in a closed fuel cycle for conversion of fertile uranium and efficient management of actinides. These reactors can operate as independent systems or in combination with thermal reactors to further utilize fissile material and transmute minor actinides accumulated in their spent uranium oxide (UOX) and MOX fuel.

Innovative thermal reactors have some specific parameters that will improve utilization of nuclear energy (e.g. high thermal efficiency or very high temperature outputs). Some of these reactors can also utilize thorium. Innovative thermal reactors will not lead to significant reductions of waste so long as the reactors operate on an open or partially closed nuclear fuel cycle; however, waste could be significantly reduced if they are deployed in combination with fast reactor systems.

Accelerator driven systems (ADSs) are innovative systems designed primarily for the final burning of plutonium and minor actinides at the end of thermal and fast reactor operations. Specific types of molten salt reactors

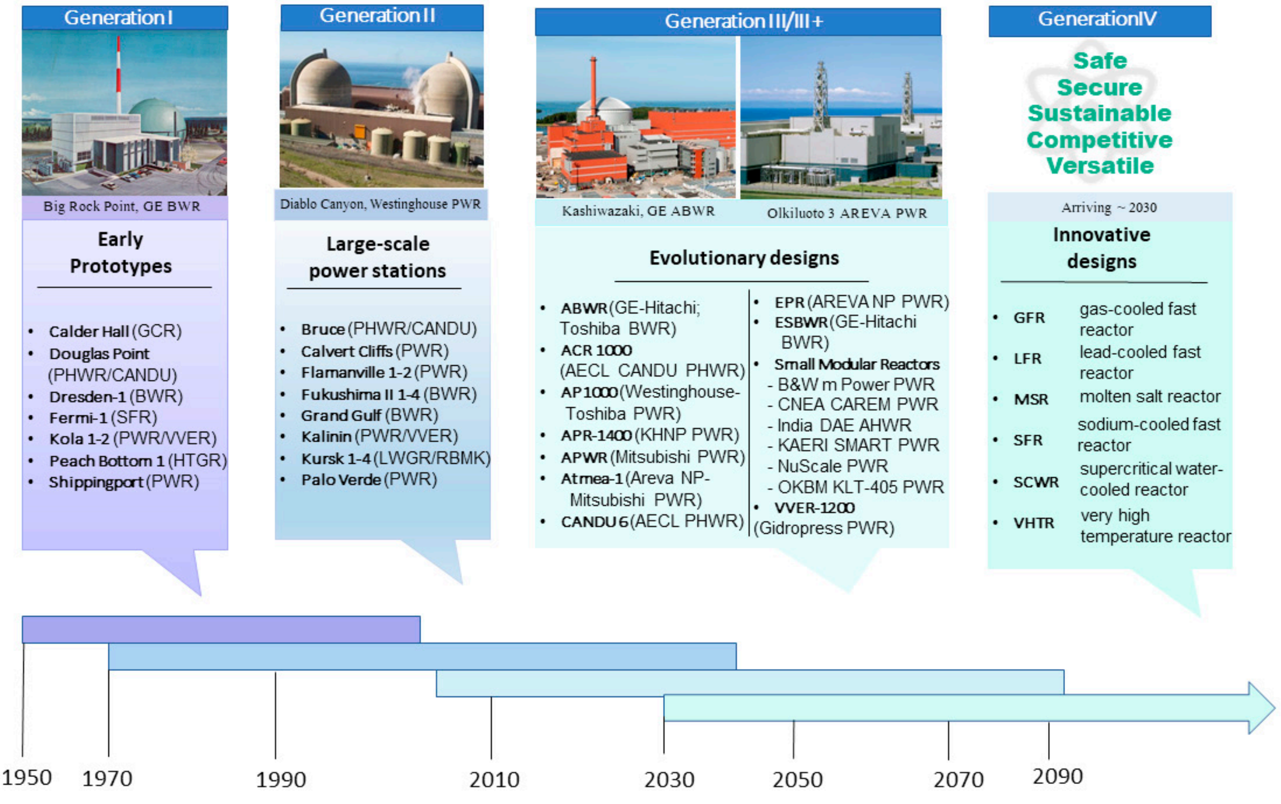

FIG. 1. Basic idea of Generation IV (A Technology Roadmap for Generation IV Nuclear Energy Systems).

(MSRs) — such as MOSART, the Russian Molten Salt Advanced Reactor Transmuter with a fast spectrum and liquid thorium fuel as its fertile element — can be dedicated to ‘burning’ (i.e. transmuting and destroying actinides).

Other reactor concepts considered in some of the innovative reactor programmes, such as small reactors and advanced light water reactors (LWRs), are not considered in this report because their waste streams are very similar to those from existing reactors and fuel cycle facilities. The treatment and disposition of these wastes is well known.

Examples of some of the reactor types are described briefly below. Further detail can be found in Annex II and the references contained therein. It is not the intent of this publication to include an exhaustive list of innovative reactor types, but to provide examples of different technologies, where such differences may have an influence on the resulting waste streams.

TABLE 1. THE BASIC TECHNOLOGIES AND APPLICABLE FUEL CYCLES

Reactor type	Neutron spectrum	Coolant	Moderator	Fuel	Fuel cycle	Reprocessing
Fast reactors						
Sodium cooled fast reactor	Fast	Na	—	U–Pu oxide carbide alloy	Closed	Aqueous Pyro
Lead cooled fast reactor	Fast	Pb Pb–Bi	—	U–Pu oxide Metal nitride	Closed	Aqueous Pyro
Gas cooled fast reactor	Fast	He	—	U–Pu carbide	Closed	Aqueous Pyro
Molten salt fast reactor	Fast	Molten fluorides	—	Molten (U or TRU–Th) fluoride	Closed	Pyro
Thermal reactors						
Very high temperature reactor	Thermal	He	Graphite	U, Th prism pebbles	Open	—
Supercritical water cooled reactor	Thermal	H_2O	H_2O	U oxide	Open	—
Advanced heavy water reactor	Thermal	H_2O	D_2O	Th–Pu Th–^{233}U oxide	Closed	Aqueous
Dedicated actinide burners						
Accelerator driven systems	Fast	Pb Pb–Bi		(U–)Pu–MA oxide (with inert matrix)	Closed (double strata)	Aqueous Pyro
Molten salt reactor	Thermal	Fluorides	Graphite	U–Pu U–Th Fluorides	Closed	Pyro

Note: —: data not available; MA — minor actinide; TRU — transuranic.

Sodium cooled fast reactor (SFR)

The SFR system is envisioned primarily to produce electricity from fissile material and excess fissile fuel from fertile ^{238}U. The operability of this system has been demonstrated at full industrial scale. Two approaches for a full actinide recycling fuel cycle are currently in development: (i) a medium to large (500 to 1500 MW(e)) SFR with MOX fuel, supported by a fuel cycle based upon advanced aqueous processing; and (ii) an intermediate size (150 to 500 MW(e)) SFR with U–Pu minor actinide zirconium metal alloy fuel, supported by a fuel cycle based on pyrometallurgical processing.

Lead cooled fast reactor (LFR)

The LFR system uses a lead or lead–bismuth eutectic liquid metal to cool the reactor. The system is in the conceptual phase of development. Its major innovative feature is the use of inert coolant to further improve reactor safety. The reactor is cooled by either natural (in small size systems) or forced (for medium–large systems) convection and sized between 50 and 1200 MW(e), with a reactor coolant outlet temperature up to 550°C (possibly ranging up to 800°C, depending upon the success of material research and development (R&D)). The LFR system could employ a long life core and effectively support an operation for minor actinide management. It is envisioned primarily for electricity production.

Gas cooled fast reactor (GCFR)

Due to the high outlet temperature of the helium coolant in a GCFR system, it can process heat or deliver electricity or hydrogen with high conversion efficiency. This system is in the conceptual phase of development. Core configurations with pin or plate based fuel assemblies are being considered, with a total core power of 300–600 MW(e).

Very high temperature reactor (VHTR)

The VHTR system can supply nuclear heat and electricity over a range of core outlet temperatures between 700 and 950°C. The original target was for an outlet temperature above 1000°C to produce thermochemical hydrogen at high efficiency and without carbon emission. Market conditions, however, showed a large market for electricity production and industrial processes based on high temperature steam with more modest outlet temperatures. So, in the past few years, the focus has changed from designs with higher outlet temperatures and direct gas turbines (such as the prismatic block fuel high temperature helium cooled reactor and pebble bed modular reactor) to lower outlet temperature designs such as the high temperature reactor pebble bed module under construction in China and the next generation nuclear plant in the United States of America. The VHTR designs are modular, with reactor powers of 200–600 MW(th). Two different fuel and core configurations are used: the pebble bed and prismatic block type. The VHTR technology is based on tristructural-isotropic (TRISO) coated particle fuel with graphite as the core structure, a helium coolant, dedicated core layout and lower power density to achieve decay heat removal in a natural way. The VHTR mostly focuses on an open, low enriched uranium fuel cycle but can support alternative fuel cycles such as U–Pu, Pu, MOX, and U–Th.

Supercritical water cooled reactor (SCWR)

The SCWR system is envisioned primarily for electricity production in an open cycle. An alternative option would involve recycling fissile material or partially burning actinides. The system uses high temperature, high pressure water (i.e. above the critical point) for cooling reactors that operate above the thermodynamic critical point of water to achieve a thermal efficiency approaching 44%. The reference plant has a 1700 MW(e) power level and a reactor outlet temperature of 550°C. The SCWR system is expected to be highly economical because of its high thermal efficiency and plant simplification. Both pressure vessel and pressure tube designs are in conceptual development.

Advanced heavy water reactor (AHWR)

The AHWR system is for electricity production in a closed fuel cycle tailored to the efficient utilization of thorium with a mix of other fissile materials. The system is light water cooled and heavy water moderated. The main part of the core is subcritical with Th–^{233}U oxide, mixed so that the system is self-sustaining in ^{233}U. A few seed regions with conventional MOX fuel will drive the reaction and give a negative void coefficient overall. In its current reference design, the AHWR utilizes a fuel assembly consisting of a combination of (Th, ^{233}U) O_2 and (Th, Pu) O_2 fuel pins. This system is in conceptual development.

Molten salt reactor (MSR)

Two molten salt reactor systems are under consideration: The first has thermal neutrons, a graphite moderator and coated particle fuel (the fluoride salt cooled high temperature reactor, or FHR), and the second has fast neutrons and liquid fuel (molten salt fast reactor (MSFR) and MOSART). The second can be configured either as a burner of plutonium and minor actinides or as a breeder using thorium as the fertile element. In this system, the fuel is a liquid mixture of fluorides containing the fissile and fertile elements, with a high boiling temperature allowing operation at low pressure (about five atmospheres). The expected fuel temperature is in the 700–800°C range. This system is in conceptual development.

Accelerator driven system (ADS)

The ADS is an advanced waste management system, focused primarily on actinide burning, that is integrated into a thermal and/or fast reactor system. ADS is based on projecting highly accelerated particles (e.g. protons) towards a heavy material target. This bombardment produces a very intense neutron source (a process called spallation) in which neutrons enter a subcritical core (also called a blanket) where they can be multiplied.

With these neutrons, Pu could be burned, a U–Th fuel cycle may be started, and very long lived radioactive isotopes such as the minor actinides can be transmuted by fission into short lived lighter isotopes. This system is in conceptual development.

2.2. INNOVATIVE NUCLEAR FUEL CYCLES

States choose to develop a nuclear fuel cycle for a variety of reasons, such as to make full use of uranium resources, minimize radioactive waste or pursue domestic energy security goals. The decision is invariably based on national policy, which varies and may include political factors as well as technical ones. If pursued, the nuclear fuel cycle should be developed in harmony with a reactor strategy in order to improve the overall economics and safety of the nuclear energy system.

Four variants of fuel cycles aimed at minimizing waste generation and decreasing resource consumption have been identified as potential reference cases for the innovative reactor concepts [1]. Figure 2(a) shows qualitatively how the required resources and the final waste products are reduced from the once through alternative to the full actinide recycling alternative, thus contributing to the sustainability of the variant. The various options are briefly described below. More detail can be found in Annex III and the references contained therein.

Note that the resource basis is not illustrating a 50-fold decrease in the need for natural uranium and thorium in the case of full recycling. Figure 2(b) (which tries to show relative amounts of waste) could be considered somewhat misleading in that it only shows a grand total of the waste to be disposed of.

The current comparison is dominated by the high volume of the uranium mining and milling waste, which is generally not a technical issue. The real issue with long lived low and intermediate level waste (L&ILW) and high level waste (HLW) is not fully noticeable due the effects of the significant decrease of uranium mining and milling waste.

An alternative approach in Fig. 2(b) shows relative amounts of the major waste categories. An open fuel cycle is shown as the base case, and other fuel cycle amounts are given relative to that (either more or less than the base case). It can be seen that the quantity of some waste types increases, while others decrease. This will change the relative sizes of disposal facilities, but will not eliminate the need for one type of facility or another (e.g. all fuel cycles will have some long lived or high activity waste that requires isolation in a deep geological repository). The differences will be in the design of the facilities, which must be tailored to suit the expected waste characteristics and amounts.

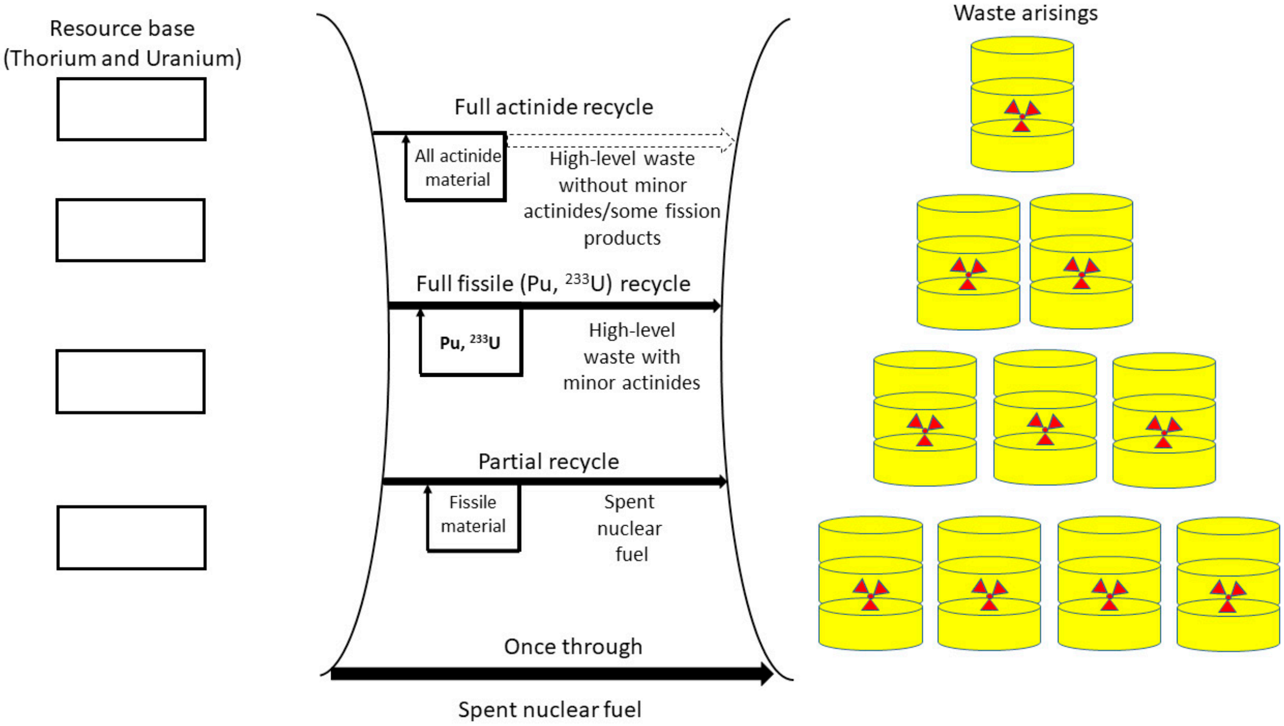

FIG. 2(a). Total waste arising from alternative nuclear fuel cycles.

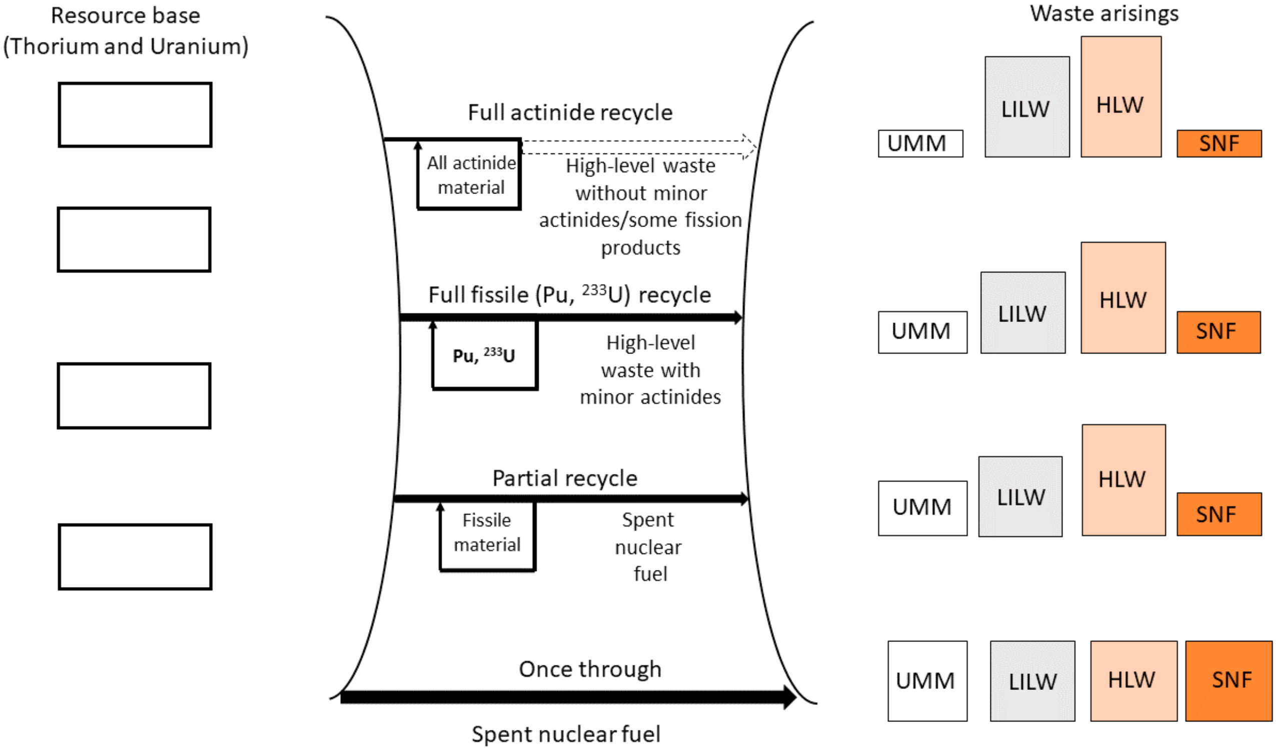

FIG. 2(b). Relative waste arising from alternative nuclear fuel cycles.

2.2.1. Current fuel cycles

Currently, there are two basic approaches to the nuclear fuel cycle: an open (or 'once through') cycle in which SNF is removed from a reactor and stored, and a closed (or partially closed) cycle in which SNF is reprocessed to produce new fuel. While existing power reactors can operate under either option, INSs may require new or modified nuclear fuel cycles.

Open cycle

For existing open fuel cycles, nuclear fuel is fabricated from uranium (or thorium), irradiated in a reactor, stored for a period of time to allow for reduction of heat output, and then directly disposed of. This is the current approach in many countries such as Canada, Finland, Germany, Sweden and the USA. Final disposal facilities for SNF do not yet exist, but are under development or consideration in several countries. Stored SNF could be retrieved for recycling purposes at any time until the final repository is sealed.

Closed or partially closed cycle

For closed or partially closed fuel cycles, actinide material is recovered from SNF in a reprocessing facility and fabricated into new fuel. The fuel is reused (generally up to three times in thermal reactors) to extract additional energy, and the resulting SNF is then disposed of as waste or stored for future applications in innovative reactors. For example, at the La Hague plant in France, low enriched uranium SNF is reprocessed, separated plutonium is fabricated into MOX fuel for further use in reactors, and the resulting MOX SNF is stored for future deployment in fast reactors. Japan, the Russian Federation and the United Kingdom also currently operate closed or partially closed nuclear fuel cycles.

2.2.2. Innovative fuel cycles

Full fissile material (Pu, ^{233}U) recycling

In a full recycling approach to the fuel cycle, all SNF is reprocessed for recovery and recycling of U, Pu and/or ^{233}U. The SNF is repeatedly processed and recycled to fully consume the fissile material through multiple irradiations in fast reactors. Minor actinides and fission products are sent to the waste stream from the processing operation. This fuel cycle supports the preservation of fissile material resources. Although this fuel cycle is not currently practiced, it could be implemented at an industrial scale with the deployment of fast reactors. For example, fuel cycles based on liquid metal fast breeder reactors have been demonstrated by France, Germany, Japan, the Russian Federation, the UK and the USA. A full fissile recycling fuel cycle could also be based on Th fuels, which would involve the recycling of ^{233}U.

Actinide and long lived fission product recycling

Another fuel cycle approach involves the recycling of actinide and long lived fission products, whereby all SNF is processed and actinides are recycled multiple times to fully consume the fissionable material and to transmute the minor actinides. One or more fission products (^{99}Tc and ^{129}I) may also be recycled. Such a fuel cycle could be based on thermal reactors (such as LWRs and/or pressurized heavy water reactors (PHWRs)) in combination with fast reactors. Both the thermal reactors and the fast reactors would produce power, but the fast reactors could also burn excess fissile fuel produced from fertile ^{238}U and minor actinides that would otherwise be sent to the repository. Concepts are being developed for such a fuel cycle for (long term) future deployment in INSs. This approach allows the preservation of resources, the minimization of the radiotoxicity of the final waste, and the optimization of repository space, which are the main criteria of innovative fuel cycles. PHWRs could also function as effective 'intermediate burners,' transmuting a significant fraction of the actinides from spent LWR fuel and significantly reducing the number of fast reactors needed for ultimate actinide transmutation. However, the implications of added fuel handling, especially of highly radioactive recycled fuels, needs to be considered, especially as it relates to worker dose and fuel quality. This may increase the complexity of fuel handling operations, requiring complex remote handling, increased waste generation, etc., and their associated costs.

Nuclear fuel cycle approaches and goals for sustainable resource utilization and waste minimization are summarized in Fig. 3.

3. WASTE ASSESSMENT GOALS AND METHODOLOGIES

Subsections 3.1 and 3.2 provide background and draw the attention of designers of innovative reactors, developers of innovative back end fuel cycles and developers of new fuels to the kind of information needed to either assess the sustainability of nuclear energy systems (NESs) or to put in place an integrated management system to deal with waste issues in all subsequent waste management steps (segregation, treatment, processing, packaging, storage and disposal), since such information has not yet been made available. Without such information, it will be much more difficult to plan required waste management facilities, to select technologies and to understand activities that need to be put in place.

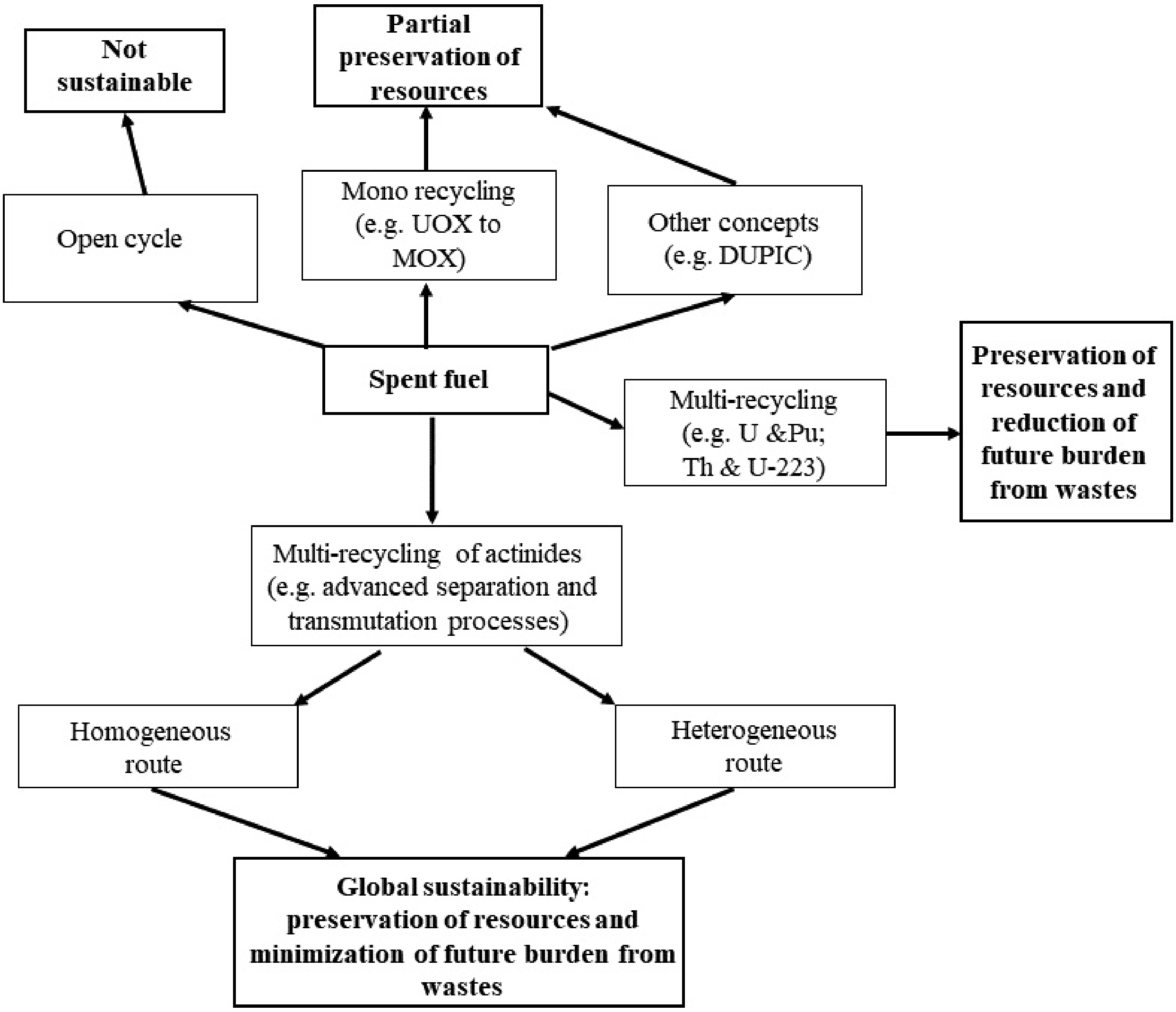

FIG. 3. Main options of the fuel cycle: From today to the future. The World Nuclear Association (WNA) defines open cycle as a sustainable fuel cycle if the waste is managed appropriately. DUPIC — direct use of [spent] pressurized water reactor fuel in CANDU reactors. Text boxes in bold represent the end result of the various options.

3.1. INPRO METHODOLOGY FOR ASSESSING SUSTAINABILITY OF NUCLEAR ENERGY SYSTEMS RELATED TO WASTE MANAGEMENT

The IAEA INPRO assessment methodology for innovative nuclear reactor systems includes an assessment of wastes and waste management issues [17]. The INPRO assessment is based on the following set of principles:

“— Generation of radioactive waste in an INS shall be kept to the minimum practicable.
— Radioactive waste in an INS shall be managed in such a way as to secure an acceptable level of protection for human health and the environment, regardless of the time or place at which impacts may occur.
— Radioactive waste in an INS shall be managed in such a way that it will not impose undue burdens on future generations.
— Interactions and relationships among all waste management steps shall be accounted for in the design of the INS, such that overall operational and long-term safety is optimized.”

The legal framework is considered part of the infrastructure. These principles are translated into a set of ‘user requirements’ and indicators, which can be assessed, measured and compared among various options. The main indicators for waste management are summarized in Table 2.

TABLE 2. INPRO WASTE MANAGEMENT ASSESSMENT INDICATORS

Indicators	Acceptance limits
Alpha emitters and other long lived radionuclides per GWa	ALARP
Total activity per GWa	ALARP
Mass per GWa	ALARP
Volume per GWa	ALARP
Chemically toxic elements that would become part of the radioactive waste per GWa	ALARP
Estimated dose rate to an individual of the critical group	Meets regulatory standards of the specific Member State
Radiological exposure of workers	
Estimated concentrations of chemical toxins in working area	
Availability of end state technology	All required technology is currently available or reasonably expected to be available on a schedule compatible with the schedule for introducing the proposed innovative fuel cycle
Time required to implement technology	Any time required to bring the technology to the industrial scale must be less than the time specified to achieve the end state
Availability of resources	Resources (funding, space, capacity, etc.) available for achieving an end state compatible with the size and growth rate of the energy system
Safety of the end state (long term expected dose to an individual of the critical group)	Meets regulatory standards of the specific Member State
Time to reach the end state	As short as reasonably practicable
Attribution of costs	Life cycle waste management costs are included in the cost estimate for the system

Note: ALARP — as low as reasonably practicable; GWa — gigawatt per year.

3.2. METHODOLOGY TO ASSESS WASTE MANAGEMENT NEEDS

The basic methodology for developing an assessment of waste management needs includes identifying current and arising waste streams, their properties, and processing methods and waste facilities infrastructure that might be needed. This methodology is applicable to both reactors and fuel cycle facilities. Figure 4 outlines the main steps in assessing waste management needs.

Consideration needs to be given to all steps of the system. Different activities in the nuclear fuel cycle will produce different types of waste, with different characteristics and requirements for further management, such as:

- Mining and milling wastes;
- Wastes from conversion, enrichment and fuel manufacturing;
- Operational wastes from NPPs;
- Spent fuel from NPPs;
- Process wastes from NPPs and from spent fuel reprocessing;
- Secondary wastes from other waste management activities (e.g. treatment of primary wastes or operation of waste management processes);
- Decommissioning waste.

All of these waste streams have to be considered in the overall assessment to develop a safe, technically sound and economical approach to waste management. In addition, a detailed plan is needed for each waste stream and for each facility in the nuclear energy system. Such a plan sets out how the waste is to be processed and managed, from generation to storage and long term disposal in a safe end state.

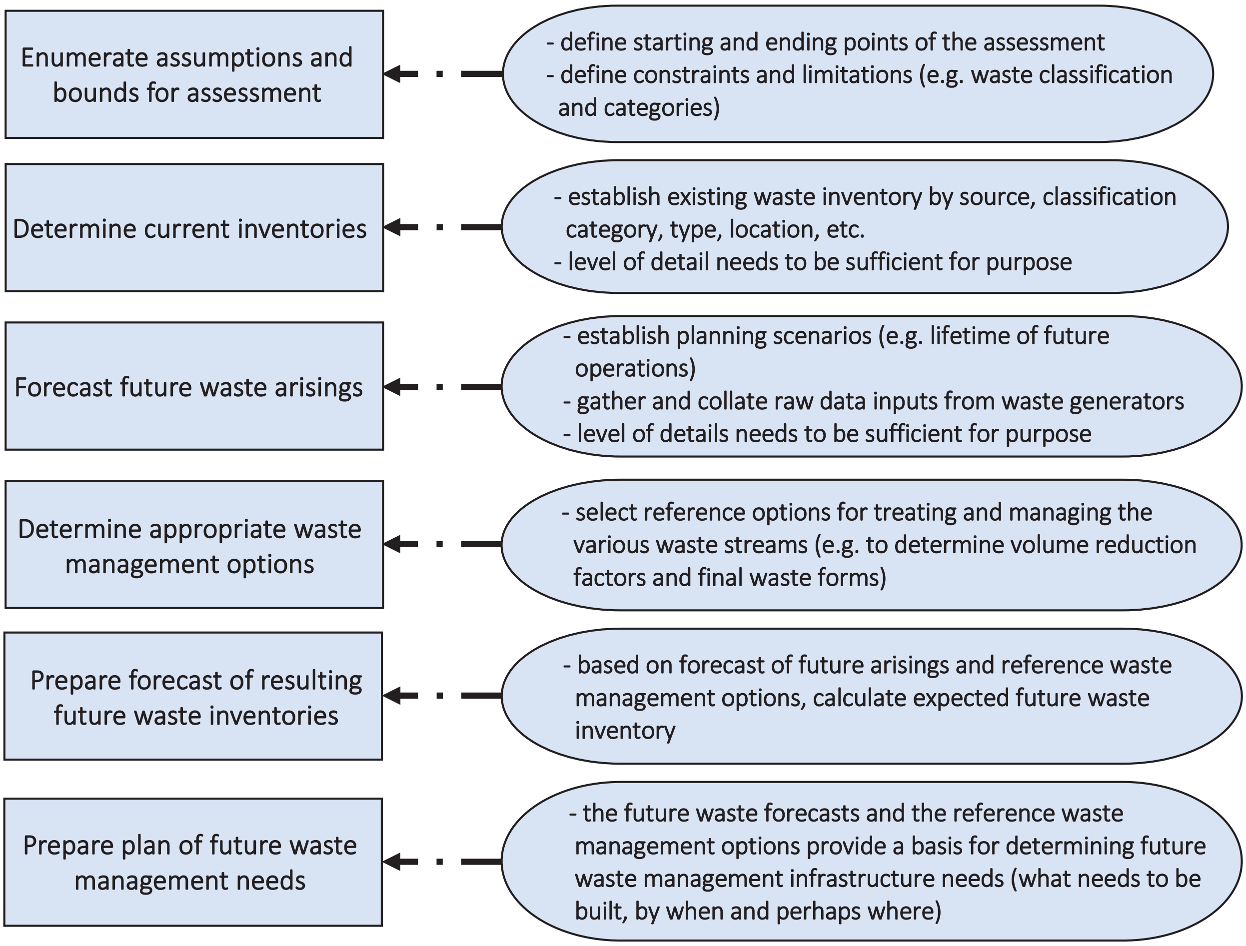

FIG. 4. Basic methodology for assessing waste management needs.

The safe end state will vary according to the waste stream, and may include:

- Recycling and reuse of the material within a nuclear or non-nuclear facility (including a possible period of storage for decay of short lived radionuclides);
- Release to the environment as a liquid or gaseous effluent;
- Near surface disposal (that limits/excludes long lived radionuclides);
- Intermediate depth or enhanced confinement disposal;
- Geological disposal (multiple barriers, including those for highly active wastes and stable waste form);
- Disposal in mine and mill tailings facilities.

Information about the waste (e.g. classification, categorization, properties and inventory) is needed in order to identify suitable waste management processes and select optimal processing technologies.

3.2.1. Waste classification

According to IAEA Safety Standards Series No. GSG-1, Classification of Radioactive Waste [18], waste can be classified as follows:

- Exempt waste (EW);
- Very short lived waste (VSLW);
- Very low level waste (VLLW);
- Low level waste (LLW);
- Intermediate level waste (ILW);
- High level waste (HLW).

This internationally accepted waste classification system classifies radioactive waste according to the activity and half-lives of radionuclides. This classification defines as acceptable, from the safety point of view, disposal routes (end points) for solid or solidified waste (plus liquid and gaseous wastes in the case of EW).

3.2.2. Waste categorization

Categorization of waste provides for consistent approaches to waste processing and storage/disposal. While classifying waste solely according to its radioactivity and half-life is a reasonable approach, it has to be complemented with additional information on the waste properties relevant for activities performed in various pre-disposal waste management steps, such as the point of origin, physical state, type of processing method, properties and process options [19–22].

Point of origin

Radioactive waste is produced from a range of activities, and the waste streams vary by form, activity, physical state and so on. The sources (point of origin) of radioactive wastes considered may include: (a) the complete nuclear fuel cycle, including the refining and conversion of uranium concentrates (yellow cake), enrichment, fuel fabrication and fuel reprocessing; (b) operation of nuclear power reactors; or (c) naturally occurring radioactive materials (NORM) waste, including uranium milling and mining.

Categorization of a waste stream based on its point of origin provides valuable insights related to expected waste stream properties. This can reduce the burden associated with subsequent characterization analyses, processes, classification and disposition.

Physical state

This is perhaps the most obvious subcategory and is comprised simply of three physical states: liquid, gaseous or solid.

Types of processing method

This subcategory is particularly useful in identifying potential treatment and conditioning technologies. In fact, waste type names are often established by the processing methods. In the case of waste to be disposed of, the waste type often relates to a portion of a repository, such as unstable or stable wastes. Examples of waste types include:

- Physical waste types (categorized for processing options based on the physical state of the waste);
- Process waste types (categorized for a known process);
- Disposal waste types.

3.2.3. Waste properties

Knowledge of waste properties assists in determining the optimal choice of process or technology that is necessary for:

- Pretreatment and treatment of primary waste;
- Conditioning for storage;
- Interim storage;
- Conditioning for disposal;
- Packaging for transport or disposal;
- Direct disposal;
- Discharge to the environment;
- Clearance.

The properties of unconditioned waste (raw, pretreated and treated) and conditioned waste (waste forms and waste packages) that need to be taken into account during the waste management process are outlined below.

Unconditioned waste properties

The three groups of unconditioned waste properties (radiological, physical–chemical and biological) are:

- *Group 1* (radiological properties):
    - Total activity and activity concentration;
    - Radionuclide composition (type of radiation, half-life);
    - Fissile mass and criticality potential;
    - Thermal power;
    - Surface dose rate;
    - Type of contamination (fixed, non-fixed);
    - Origin of the activity (contamination or activation).
- *Group 2* (physical properties and chemical properties):
    - Physical state;
    - Volume, mass and dimensions of waste items;
    - Density;
    - Volatility;
    - Chemical composition;
    - Combustibility and thermal resistance;
    - Chemical compatibility;
    - Ignitability, pyrophoricity;
    - Gas generation;
    - Acidity/alkalinity (pH);
    - Toxicity.

- *Group 3* (biological properties):
  - Putrescibility;
  - Infectious/pathogenic.

Properties of waste forms and waste packages

The two groups of conditioned waste properties (radiological and physical–chemical) that provide the bases for acceptance criteria for waste storage or disposal are:

- *Group 1* (radiological properties):
  - Total activity;
  - Radionuclide composition;
  - Surface dose rate;
  - Surface contamination;
  - Thermal power;
  - Radiation stability;
  - Fissile content.
- *Group 2* (physical and chemical properties):
  - Mass and weight;
  - Structural and dimensional stability;
  - Permeability and porosity;
  - Density;
  - Voidage;
  - Mechanical strength/load resistance;
  - Impact resistance;
  - Homogeneity;
  - Chemical stability (leachability);
  - Chemical composition;
  - Corrosivity;
  - Explosiveness;
  - Gas generation;
  - Toxicity;
  - Thermal stability;
  - Fire resistance.

3.2.4. Waste inventory

A waste inventory summarizes the knowledge of waste generators, waste processors and waste disposal operators about current or forecasted waste streams. It includes information about classification, categorization, and properties of individually defined waste streams, as well as the timescale and dynamic of waste production. This information is needed to manage different waste streams; determine the necessary technologies, processes or resources; and establish the timing needed to develop an adequate infrastructure. On the other hand, some information cannot be obtained at the design stage since it needs to take into account the operating experiences related to maintenance, upsets and outages, and decommissioning practices for reactors and nuclear fuel cycle (NFC) facilities.

3.3. MANAGEMENT OF WASTE

Intermediate waste management steps should ensure that waste packages fulfil the waste acceptance criteria of the safe end state. Secondary waste and gaseous or liquid emissions from waste processing facilities need to be considered when evaluating waste conditioning methods.

The most desirable approach for reducing waste is to do so at the source. However, reduction at the source is limited by the need to operate effectively, economically, and sustainably. The waste that is produced can be treated to reduce the volume requiring disposal. Such reduction is already achieved in many facilities using current technologies, including:

— Compaction, super compaction, incineration, sintering and melting (for solids).
— Chemical precipitation, evaporation, ion exchange and membrane separation (for liquids).
— Thermal solidification of liquid concentrates (bituminization, vitrification, drying).

New technologies for volume reduction are also being investigated, such as:

— Cold crucible melting and plasma melting;
— Non-flame technologies such as steam reforming, electron beam, ultra violet photo-oxidation and supercritical water oxidation.

These techniques can be applied or adapted to most waste types that are expected to be created during the operation and decommissioning of the innovative NPPs and reprocessing of spent fuel, once any special pretreatment or other considerations have been taken into account (e.g. removal of sodium from sodium wetted solid waste). Some waste types, such as lead based coolants, molten salts and liquid fuels, will require additional research to create and demonstrate waste management options and waste forms.

3.4. METHOD TO COLLECT AND STRUCTURE INFORMATION ON WASTE FROM INNOVATIVE REACTORS AND FUEL CYCLES

There are many potential waste streams from a variety of sources that need to be considered in an innovative fuel cycle, as indicated in Fig. 5.

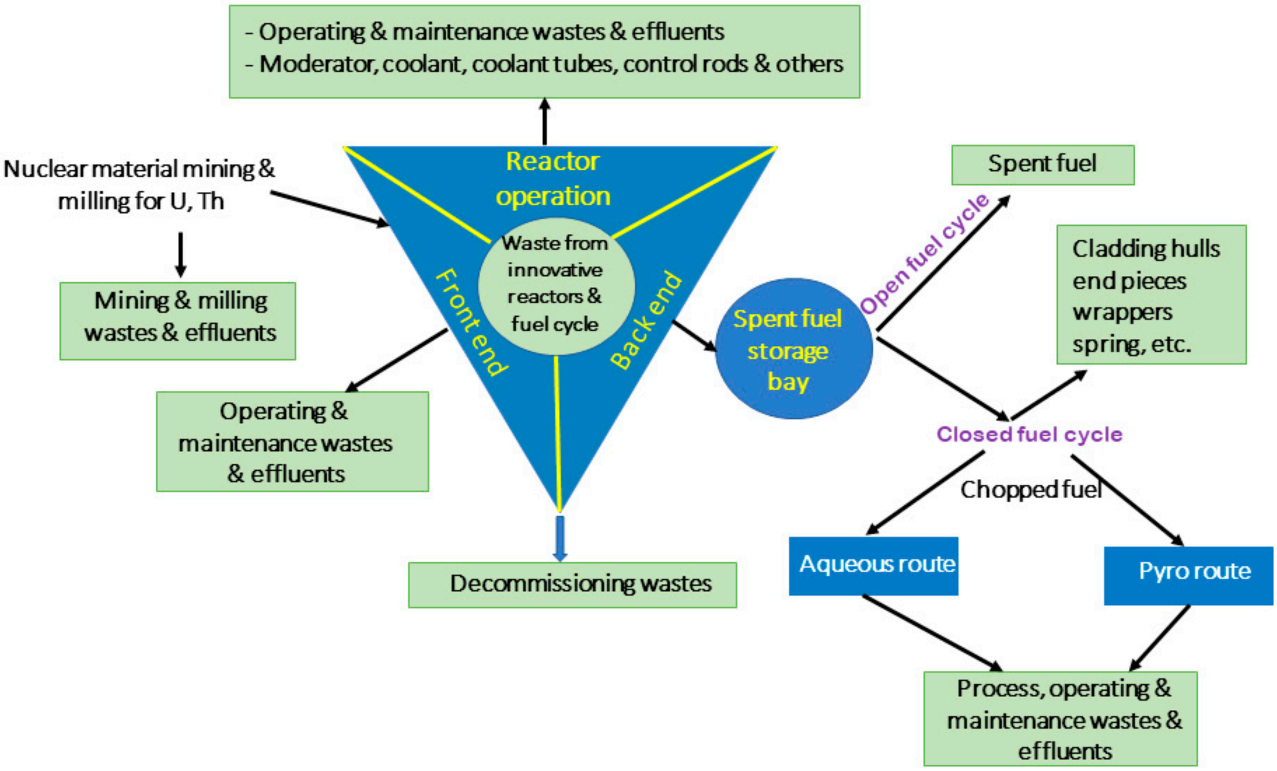

FIG. 5. *Examples of waste sources in a typical nuclear fuel cycle.*

As most of the innovative nuclear reactor and nuclear fuel cycle systems are in development, it is difficult to provide detailed estimates of the amounts and characteristics of the wastes that they will produce. Therefore, a systematic approach to collecting and structuring information relevant to future management of waste from innovative reactors and nuclear fuel cycles needs to be outlined early in development, in order to:

- Assess the sustainability of these innovative concepts from the waste management perspective (e.g. INPRO methodology);
- Assess whether challenges related to waste management may impact timely deployment;
- Identify problematic waste streams that may require either further enhancement of existing waste management processes or technologies, or initiation for development of a new one;
- Identify and eventually quantify principal waste streams that are similar to the waste streams of the current NPPs and NFCs and the options for the life cycle management (e.g. processing and storage) of each waste stream, up to and including final disposition (e.g. repository).
- Identify, sort and segregate techniques which may lead to recycling and reuse opportunities that reduce the radioactive waste volume.

Collecting and structuring data on waste streams from innovative concepts is an iterative effort that will have to be performed in different phases of development and deployment. For example, at the design and development stage there will be little data on waste quantities and characteristics. Therefore, initially, the assessment needs to be performed on a theoretical, case by case basis. Knowledge of the process flowsheet for each step in the cycle, as well as interactions between each step, material balances, and construction materials, among other things, are essential for further estimating waste types, quantities and characteristics. Additional information will have to be obtained from experiences with similar designs and/or laboratory scale designs.

However, the information that would become available just prior to construction will have to be significantly more detailed — as detailed as available information from current reactors and fuel cycles — to allow a full sustainability assessment and a more detailed assessment of waste management needs.

To promote dialogue between experts in waste management and the designers and developers of innovative concepts, information for the first assessment of waste streams from innovative reactors needs to be collected and structured. An approach to structuring this information should include the following independent steps:

- Identification of principal waste streams from operation and decommissioning of innovative reactors, including challenges and problematic waste streams;
- Identification of principal waste streams from the innovative nuclear fuel cycle facilities, including challenges and problematic waste streams;
- Identification of probable waste streams, including wastes foreseen from ultimate decommissioning of innovative reactors and nuclear fuel cycle facilities;
- Review of the total waste expected and waste management issues identified for each type of innovative facility;
- Development of recommendations on priorities for addressing challenges, problematic streams and suggestions for further studies, as well as the level of detail of missing data on waste streams relevant to envisaged waste management practices.

4. WASTE FROM INNOVATIVE REACTORS

4.1. GENERAL APPROACHES

The designers of the innovative reactors considered in this publication have proposed a number of recommendations for achieving waste management goals. They include:

- Operating at very high fuel (uranium) burnup in an open fuel cycle and/or as a breeder or near breeder: In a closed fuel cycle, the volume of waste fuel per kilowatt-hour produced by the reactor must be reduced.

- Improving the efficiency of the energy conversion process: reduces the waste produced per unit of energy to the end user.
- Operating on a closed fuel cycle with recycling of minor actinides: Actinides are recovered during reprocessing and returned to the reactor where they are consumed.
- Using thorium as a major component of the reactor fuel: reduces the quantity of higher actinides produced in the reactor (e.g. plutonium).
- Improving the utilization of uranium and thorium resources: More efficient use of fertile material in the reactors would decrease the resources needed and reduce the need for and impacts of uranium and thorium mining and milling.

4.2. DESIGN MEASURES

The design stage offers the greatest potential for reducing waste, as it offers the maximum flexibility to adjust the characteristics of the system for this purpose. The minimization of waste by design is inherently safer than depending upon operational practices. It is particularly important to reduce components of the waste that are radiologically and chemically toxic for a long time and that are mobile in the repository environment. Design measures aimed at reducing the generation of radioactive waste in existing nuclear energy systems are well known and described [23]; these measures should also be applied during the design of innovative reactors. Particular attention should be paid to:

- Building and equipment layout;
- Selection of components (including consideration of construction materials);
- Application of advanced materials, such as cobalt free steels to reduce activation;
- Reducing surface contamination;
- Reducing leakage and crud traps;
- Limiting corrosion;
- Minimizing the spread of contamination;
- Preplanning for decommissioning, such as provisions to facilitate and improve decontamination, dismantling and segmentation;
- Advanced methods of waste treatment and disposal;
- Minimum use of hazardous materials;
- Reducing secondary waste generation.

There are a large number of specific designs currently being considered for innovative reactors. Each of these will have different construction and operating characteristics, which will affect the precise nature and quantity of waste produced by the reactor and its associated fuel cycle. Since most of the innovative reactors are still in the conceptual design stage, it is only possible to indicate what waste streams could be expected. Comparison at the present stage can only be made without a detailed quantitative assessment of volumes and activity. As a result, proposed INSs need to be evaluated on a case by case basis. Using a standard methodology allows different systems or alternative variations to be compared on an equal footing.

4.2.1. Waste from advanced water cooled reactors

The following technical features to minimize the generation of waste are commonly mentioned by the designers of innovative water cooled reactors such as SCWRs and AHWRs:

- Soluble boron-free core.
- Simplification of the primary circuit and reduction of the equipment achieved by, for example, the use of integral design, compact design or minimized connections and piping, resulting in a reduced amount of activated or potentially contaminated structural materials (all innovative water cooled reactors).
- Increased operational interval, contributing to the reduced volume of high level waste. (Note however, that reduction in waste volumes may also lead to an increase in specific activity of some wastes, which

may cause complications in the waste management system. Volume and activity considerations need to be carefully balanced.)

Infrequent refuelling or automated on-power refuelling, integral design of primary circuit and reduced piping are also specified as factors contributing to the minimization of dose rates achieved through the resulting reduced maintenance.

To achieve improved fuel utilization and to broaden the available base of natural resources, the designers of many innovative water cooled reactors foresee, as an alternative, operation within a partially closed fuel cycle or the introduction of thorium fuel (in AHWRs). AHWRs achieve an increased conversion with the use of the Pu–Th fuel and on-line refuelling, and also offer minimized production of the highly radiotoxic long lived minor actinides. However, they produce a large quantity of toxic ruthenium. The designers of the AHWR also consider zirconium recycling.

The thorium fuel cycle serves to reduce the quantity of spent fuel that contains the long lived fission products of the higher uranium isotopes. Fission of ^{233}U (produced from ^{232}Th) does not yield the same longer lived minor actinides associated with the higher uranium isotopes. This could reduce the cost of spent fuel disposal. The low and intermediate level waste volumes generated by a water cooled reactor operating on the thorium fuel cycle would be similar to those of a water cooled reactor of the same size and design (e.g. pressurized water reactor (PWR) or PHWR) operating on a conventional uranium based fuel cycle.

4.2.2. Waste from gas cooled reactors

For gas cooled reactors (VHTRs and GCFRs), the features commonly mentioned in conjunction with effective waste minimization and reduced environmental impacts are:

- High efficiency of energy conversion (e.g. achieved through the use of direct or indirect Brayton cycles and through purposeful use of the rejected heat);
- High fuel burnup that, together with efficient energy conversion, contributes to a more effective utilization of uranium as compared to presently operated LWRs (in the case of VHTRs);
- A proven radiological cleanness of the coated particle fuel, resulting from the proven perfect confinement capability of such fuel at high temperatures;
- Reduced water consumption and improved land use allow utilization of a dry cooling system;
- Reduced HLW, resulting from a high degree of depletion in the spent fuel in comparison with the once-through LWRs (in the case of VHTRs);
- The absence of waste in the form of activated metals (e.g. irradiated claddings);
- An option to use thorium fuel and an option to operate in a closed fuel cycle for GCFRs.

Radioactive releases from the GCFR during operation are lower due to the retention properties of the graphite and the fact that the chemically inert helium coolant is not activated. Actinide production is estimated to be lower than that of a PWR (on a per megawatt basis) if the VHTR utilizes thorium, which would reduce the amount of long lived actinides to be managed in the waste. The graphite itself can become a problematic waste, potentially requiring disposition in a deep geological repository. Also, decommissioning waste can be higher by volume than LWR waste because of the much lower power density and the larger physical size of the reactor per MW installed.

4.2.3. Waste from liquid metal cooled reactors

For liquid metal cooled reactors (sodium, lead and lead–bismuth), features of waste minimization and reduced environmental impact are commonly mentioned in conjunction with their use in a closed fuel cycle. For example:

- High conversion or fuel breeding and operation in a closed nuclear fuel cycle can ensure self-sustaining operation (with the breeding ratio BR ~1.05) or expanded breeding (BR >1) to produce fissile materials for other, non-breeder reactors in the system.

- While all reactors with the fast neutron spectrum generally feature high conversion or breeding, performance can be improved by the use of dense metallic or nitride fuel (e.g. lead or lead–bismuth reactors). For the nitride fuel, enrichment by ^{15}N is being considered.
- Options to recycle all self-produced transuranic elements (TUE) (including minor actinides) or to utilize SNF from other reactors (e.g. LWRs) are being examined for both above mentioned approaches. This would result in the reduction of long lived waste. Specific reduction targets will depend on the technology and recycling option chosen.

All innovative liquid metal cooled reactors foresee operation in a closed fuel cycle involving advanced methods of fuel reprocessing, with the potential to reduce waste generation.

Note that the chemical toxicity of lead and/or bismuth coolants is a concern for their long term management, especially as it relates to possible long term leaching into the environment from disposal facilities.

4.2.4. Waste from dedicated actinide burners — molten salt reactors

A unique feature of the liquid fuelled MSR is that the fertile (mainly ^{232}Th) and fissile (^{233}U/^{235}U/^{239}Pu) isotopes are dissolved in a fluoride based molten salt that functions as both the fuel and the coolant. When fast neutrons are used, this type of reactor can burn plutonium and minor actinides. Because the presence of thorium in the fuel salt or in a blanket salt allows regeneration of ^{233}U, it can be used as a breeder or a burner. Therefore, the liquid fuelled MSR could support the transition between the presently common U–Pu open cycle and a future Th–U closed cycle; most of the actinide load would be burned by the end of operation of the Th–U cycle. The MSR is also expected to have a strong safety performance due to its liquid fuel — which prevents significant reactivity reserves — strongly negative feedback coefficients, or passive draining of the fuel salt into a non-critical configuration in case of emergency.

The reactor operation wastes are mainly divided into two streams: noble fission products that are extracted on-line during operation, and fission products that remain in the fuel salt (mainly rare earths) and are extracted by a pyrochemical reprocessing of the fuel at a small rate (on the order of 10 L/d/GW(e)). The pyrochemical reprocessing of the fuel salt is compulsory for such reactors. Thus, a solution must be developed for safely handling the salts, and for separating the actinides and fission products from the base salt. Several flowsheets for this are being developed, but a large number of experimental studies are needed to reach laboratory scale feasibility. Waste forms and packages will need to be developed and demonstrated to contain the highly corrosive nature of some salt materials, such as fluoride salts.

4.3. WASTE STREAMS AND CHARACTERISTICS

Although it is difficult to compare wastes from innovative reactors without their detailed designs, it is worthwhile to investigate potential issues in waste form or waste treatment by making a qualitative comparison of the low and intermediate level (L&IL) waste streams envisioned in conceptual designs.

There are six main families of innovative reactors under development: sodium cooled, lead cooled, gas cooled, very high temperature, water cooled and MSR. These reactors will use five kinds of fuel chemical forms: oxide, carbide, nitride, metal and molten salt. Tables 3–8 summarize the main waste streams from operation and decommissioning of each of the innovative reactor types. The focus is on ‘new waste streams’ or ‘unique wastes’ which are different from waste streams encountered at current water or gas cooled nuclear power reactors, along with any expected issues that may arise from the management of these waste streams.

These reactor types have not yet been fully designed and selection of materials for various systems has not been finalized, so it is not possible to estimate the quantity and characteristics of every waste stream. However, where information exists for similar, existing reactor concepts, it has been used as a basis for the summary. New construction materials for the innovative reactor systems, along with differing neutron spectra, may produce different radionuclides during neutron activation. These radionuclides will eventually be found in the various waste streams. In some cases, their behaviour in waste forms is unknown and needs to be researched.

In general, ‘routine wastes’ include the solid, liquid and gas waste streams that are similar to current waste streams, such as the residue on protective clothing, decontamination liquids, process stream filters, replaced equipment and other such wastes. Processing, conditioning, storage and disposal of these wastes are routine practices today. Improved methods may be developed in future years, but new techniques would only represent an evolutionary improvement over the current methods, rather than completely new techniques developed for completely new waste types.

Tables 3–8 are intended to set out a systematic starting point for collection of waste stream data for discussions with reactor designers. While it is recognized that waste management issues may not be a primary driver in the design of the reactor systems (e.g. in comparison to the need for systems to be able to withstand the harsh radiological and chemical conditions of some of the reactor concepts), the design and choice of materials will have an impact on waste management requirements.

These tables also focus on identifying potentially problematic wastes [24, 27] broadly defined as wastes which are different in composition or character than wastes produced by current reactor systems, or for which there is no existing management route. Where available, the tables also contain cross references to other publications dealing with these wastes.

The tables that summarize waste streams in Section 4 do not include nuclear fuel. For open fuel cycles, SNF may be considered waste. For closed fuel cycles, wastes from nuclear fuel reprocessing and associated processes are considered in Section 5.

If nuclear energy expands and continues to operate for hundreds of years, recycling and reuse of fissile and fertile material will become a necessity. Elimination of residual long lived radiotoxic nuclides would reduce the radiotoxic inventory and its burden on future generations.

Reprocessing and recycling may or may not include processing and treatment of the actinides and long lived fission products. If so, the long lived radionuclides would be partitioned from HLW, some of which would be transmuted while others could be used or separately disposed of. Without transmutation, the separated long lived actinides could be immobilized in a tailored ceramic or other matrix, which may have advantages over vitrification.

4.4. GENERAL APPROACHES

Improved approaches to the long term safety of radioactive waste management should lead to reductions of radiotoxicity, heat load and waste to be disposed of. General approaches of the innovative fuel cycles summarized in Fig. 3 are described here in more detail. Note that approaches to long term safety should lead to a reduction of long lived nuclides that have an impact on dose. These are not the same as the ones considered to be radiotoxic (e.g. ^{129}I and ^{36}Cl vs. heavy metals, which are very effectively retained in waste forms and geospheres).

The radiotoxicity in the spent fuel is defined by the presence of fission products and actinides [28]. The evolution of spent fuel is very well known and depends on the type of fuel and the attained burnup. During the first years, the fission products ^{137}Cs–(Ba) and ^{90}Sr–(Y) determine the radioactivity and heat emission. From several decades to a period of about 250 000 years, plutonium and growing americium isotopes are the main contributors to radiotoxicity. Beyond 250 000 years, ^{237}Np emerges as a main contributor, together with the progeny of uranium.

The recycling of plutonium into LWR MOX fuel generates a significantly more radiotoxic spent fuel. Typically, this fuel is sent to interim storage facilities to await a future decision regarding its final disposal or further use as fuel for innovative fast reactor systems, should they become economical [29]. The radiotoxicity of this fuel comes from the cumulative decay curves of ^{244}Cm, ^{238}Pu, ^{241}Am, ^{239}Pu, ^{240}Pu and ^{242}Pu. The other alpha nuclides (e.g. ^{243}Am, ^{245}Cm and ^{237}Np) are negligible in this context.

The alpha radiotoxicity of fast reactor mixed U–Pu fuel (whether in metallic, oxide, nitride or other form) is approximately two times higher than that of LWR MOX, taking into account the increased plutonium concentration (~8–22%) in the fuel of a number of fast reactors that have been operating over several decades (DFR, PFR, Phénix, Superphénix). But the radiotoxicity expressed per unit of energy produced is nearly the same [28] irrespective of the burnup level. An advantage of the fast reactor fuel cycle is that the fast neutron spectrum increases fission yields of even and odd actinides and then transmutes all alpha nuclides into shorter lived fission products (FPs), decreasing the alpha radiotoxicity of the spent fuel. Alpha radiotoxicity continues to increase in an LWR MOX fuel cycle.

In a partially closed cycle, only uranium and plutonium are separated from the spent fuel and recycled into fresh fuel (e.g. MOX fuel). The minor actinides are diluted in the high level liquid waste (HLLW) with the fission products and conditioned by vitrification, then dispersed in the volume of HLW while in repository. In a

TABLE 3. SUMMARY OF WASTE FROM SODIUM COOLED FAST REACTORS

Parameter	Reactor operations[a]				Decommissioning			
	Routine waste	Unique waste			Routine waste	Unique waste		
Description	Solid, liquid and gaseous waste from operation	Sodium wetted combustible waste	Sodium wetted metallic waste	Bulk sodium from cooling system components	Dismantled components and structures	Sodium wetted combustible waste	Sodium wetted metallic waste	Bulk sodium from cooling system
Type	L&ILW	L&ILW	L&ILW	L&ILW	L&ILW	L&ILW	L&ILW	L&ILW
Environmental impact	Minimal	Sodium will react violently with moisture and air, representing a significant fire hazard. Reactions will also form NaOH, which is highly caustic.			Minimal	Sodium will react violently with moisture and air, representing a significant fire hazard. Reactions will also form NaOH, which is highly caustic.		
Existing processing/ disposal available	Various	Various processes have been developed to treat the sodium, such as controlled reaction with NaOH, transformation to sodium carbonate, water vapour nitrogen (WVN), etc.			Various	Various processes have been developed to treat the sodium, such as controlled reaction with NaOH, transformation to sodium carbonate, water vapour nitrogen (WVN), etc.		
Potential problems	Care must be taken to ensure sodium is removed or neutralized	Sodium must be removed or neutralized (e.g. controlled conversion to NaOH) prior to treatment or storage of the underlying wastes.			Care must be taken to ensure sodium is removed or neutralized	Sodium must be removed or neutralized (e.g. controlled conversion to NaOH) prior to treatment or storage of the underlying wastes. Each reactor may contain several thousand tons of sodium coolant.		
Comments	Readily available and well understood processes	See Ref. [24] for details of various processes.			Readily available and well understood processes	See Ref. [24] for details of various processes. Fuel is assumed to be removed prior to decommissioning.		

Note: L&ILW — low and intermediate level waste.

[a] Does not include nuclear fuel.

TABLE 4. SUMMARY OF WASTE FROM GAS COOLED FAST REACTOR

Parameter	Reactor operations		Decommissioning	
	Routine waste	Unique waste	Routine waste	Unique waste
Description	Solid and gaseous waste from operation	Ceramic waste	Dismantled components and structures	
Type	L&ILW	L&ILW	L&ILW	L&ILW
Environmental impact	Minimal			
Existing processing/ disposal available	Various	Long term storage/ disposal	Various	Long term storage
Potential problems	No significant problems expected; substantial international experience with routine GCR wastes		No significant problems expected; substantial international experience with routine GCR wastes	
Comments	Readily available and well understood processes [a]		Readily available and well understood processes	

Note: GCR — gas cooled reactor; L&ILW — low and intermediate level waste.

[a] See Ref. [25] for details of various processes.

TABLE 5. SUMMARY OF WASTE FROM LEAD COOLED FAST REACTORS

Parameter	Reactor operations				Decommissioning			
	Routine waste	Unique waste			Routine waste	Unique waste		
Description	Solid, liquid and gaseous waste from operation	Lead wetted combustible waste	Lead wetted metallic waste	Bulk lead from cooling system components	Dismantled components and structures	Lead wetted combustible waste	Lead wetted metallic waste	Bulk lead from cooling system components
Type	L&ILW	L&ILW	L&ILW	L&ILW	L&ILW	L&ILW	L&ILW	L&ILW
Environmental impact	Environmental risk associated with lead/lead–bismuth toxicity is mitigated by control of the lead processing (at 400°C the lead concentration in the atmosphere is well below the limits for exposed workers). Standard provisions set forth by the conventional lead industry can be used.				Environmental risk associated with lead/lead–bismuth toxicity is mitigated by control of the lead processing. Standard provisions set forth by the conventional lead industry can be used, but may need to be modified to take into account the radioactive nature.			
Existing processing/ disposal available	Various[a]	Various simple and effective processes already developed and successfully implemented in European lead laboratories			Various[a]	Various simple and effective processes already developed and successfully implemented in European lead laboratories		Solidification and confinement
Potential problems	Care may be required when handling lead and bismuth contaminated materials (e.g. controlling the temperature and the formation of aerosols).				Care may be required when handling lead and bismuth contaminated materials (e.g. controlling the temperature and the formation of aerosols. Restrictions on lead and bismuth disposal may create issues for disposal).			
Comments	Readily available and well understood processes	Readily available and well understood processes used till now in small scales need to be developed for a large scale. No specific concern.			Readily available and well understood processes	Readily available and well understood processes used till now on a small scale need to be developed for a large scale. No specific concern.		The bulk lead might be confined (solid) and used in another reactor

Note: L&ILW — low and intermediate level waste.

[a] Cleaning Methods for LFR/SFR Components, ENEA Report NNFISS, LP3 – 021 2/9/2011 (public, in Italian).

TABLE 6. SUMMARY OF WASTE FROM MOLTEN SALT FAST REACTORS

Parameter	Reactor operations		Decommissioning	
	Routine waste	Unique waste	Routine waste	Unique waste
Description	Solid and gaseous waste from operation	Activated or fuel contaminated metallic and carbide waste. Salt waste from cleaning operations.	Dismantled components and structures	Bulk salts from the cooling system
Type	L&ILW	L&ILW	L&ILW	L&ILW
Environmental impact	Minimal			
Existing processing/ disposal available	Generic concept for waste treatment. Detailed processes have to be investigated.		← idem	
Potential problems	Gaseous hydrogen fluoride might be produced by fluoride hydrolysis. Fuel salt traces are removed by washing in a clean fluoride bath before waste recovery. The resulting contaminated salts need to be treated with the same type of pyrochemical process as the fuel salt is.		← idem	
Comments	Routine and unique wastes treatments share some common processes with fuel treatment. These steps are assumed to be well acquired before reactor building at the industrial scale. However, it is too early to draw any definitive conclusions since reactor design and material selections have not been finalized.		← idem	

Note: L&ILW — low and intermediate level waste.

TABLE 7. SUMMARY OF WASTE FROM SUPERCRITICAL WATER COOLED REACTOR

Parameter	Reactor operations		Decommissioning	
	Routine waste	Unique waste	Routine waste	Unique waste
Description	Solid, liquid and gaseous waste from operation		Dismantled components and structures	
Type	L&ILW		L&ILW	
Environmental impact				
Existing processing/disposal available	Various		Various	
Potential problems	Routine waste should be similar to waste from other water cooled reactors.		Routine waste should be similar to waste from other water cooled reactors.	
Comments	Readily available and well understood processes		Use of exotic alloys and other materials in core areas may result in the production of significant quantities of long lived radionuclides (e.g. certain transition metals). This may also give waste volume reduction concerns.	

Note: L&ILW — low and intermediate level waste.

TABLE 8. SUMMARY OF WASTE FROM VERY HIGH TEMPERATURE REACTORS

Parameter	Reactor operations			Decommissioning		
	Routine waste	Unique waste		Routine waste	Unique waste	
Description	Solid, liquid and gaseous waste from operation	Graphite prismatic skeletons of fuel assemblies	Tritium water	Dismantled components and structures	Graphite	Spent fuel
Type	L&ILW	L&ILW	ILW	L&ILW	L&ILW	HLW
Environmental impact	Minimal					
Existing processing/ disposal available	Various	Long term storage	Long term storage	Various	Long term storage	Long term storage/final disposal/ reprocessing
Potential problems	No significant problems expected. Substantial international experience with routine GCR waste.	Large quantity of spent graphite	Tritium will be collected through a helium purification system during operation	No significant problems expected	Large volume, containing ^{14}C	Benefit for reprocessing is less, technology for reprocessing is less, volume for final disposal is large if not separated from the matrix graphite[a]
Comments	Readily available and well understood processes	Cost effective technology required to reduce volume of graphite[a]		— Cost effective technology required to reduce volume of graphite[b]; — Use of exotic alloys and other materials in core areas may result in production of significant quantities of long lived radionuclides (e.g. certain transition metals); — Issues associated with disposal of bulk graphite (e.g. Wigner energy); — Coated particle fuel is already in an ideal form for long term storage or disposal with high level of retention/immobilization and a much lower heat production density[c].		

Note: GCR — gas cooled reactor; L&ILW — low and intermediate level waste.

[a] Since the burnup is 80–200 GW·d/te, the actual volume of HLW will most probably be smaller depending on what waste minimization is done (such as separating coated particles from the graphite matrix and thus, by implication, out of the block or pebble).

[b] See Ref. [25] for details of various processes.

[c] See Ref. [26] for more details on waste from innovative nuclear fuel cycles.

closed cycle, all actinides are separated from the spent fuel and recycled in the fast reactors by homogeneous or heterogeneous routes. The HLW contains only fission products (and actinides from process losses) in a repository, so the radiotoxicity will be low after hundreds of years. However, it is important to note process losses (which may be up to about 5%), which will result in some concentration of actinides in the HLW and or ILW. The inventory of actinides will be present on the surfaces inside reactors and fuel cycle facilities, and they will increase over time as they are processed, stored and cycled to fast burner reactors or ADS.

The radiotoxicity of target fuel in fast burner reactors and accelerator driven transmutation systems is an order of magnitude higher than LWR MOX fuel (up to 42% plutonium and a burnup of 210 GW·d/t HM), which could present a safety concern. The safety of the fuel cycle, reactors and interim storage systems could be affected by the different chemical and physical nature of the materials containing the actinides. In the context of reducing the radiological risk for a deep geological repository, the long lived water soluble fission products ^{129}I, ^{135}Cs, ^{79}Se and ^{126}Sn are the most important radionuclides, due to a combination of toxicity, half-life and concentration. The most toxic of the fission products is ^{129}I, which has a specific toxicity (Sv/Bq) similar to that of the actinides. The other soluble fission products are less toxic by two or three orders of magnitude compared with the actinides [28]. Some fission products (e.g. ^{99}Tc and ^{107}Pd) have long half-lives but are only slightly soluble in the chemical reducing media representative of most deep geological repository conditions. The solubility of ^{99}Tc (as TcO_4) in oxidizing conditions (e.g. Yucca Mountain in the USA) is much higher.

An advanced fuel cycle with partitioning followed by ‘improved’ conditioning of selected radionuclides would substantially decrease the migration risk, but not the (potential) hazard. Separation of minor actinides (MAs) before vitrification and special conditioning of these radionuclides into, for example, ceramic or crystalline matrices with a very low solubility in water is a possibility that would offer substantial advantages in the reduction of long term migration risk. However, such practices do not reduce the radiotoxic inventory and its associated hazard. The reduction of the actinide inventory, or its mean half-life, is the only possibility for significantly reducing the long term hazard. Only nuclear processes that are capable of modifying the nature of the isotopes involved and their associated half-lives or decay schemes can achieve such a goal, as elaborated below.

The largest hazard reduction can be obtained through the fissioning (transmutation) of the higher actinides, which decreases the intrinsic radiotoxicity by a factor of 100–1000 [28]. This nuclear process is preferably carried out with fast neutrons, if available in excess [28]. (Note that minor actinide transmutation is very effective in thermal reactors and can be used in place of current burnable poisons. Plutonium requires fast neutrons if it is to be fully transmuted. A few passes for plutonium can be used in thermal reactors before going to fast reactors, although not many.)

Among various strategies for the nuclear fuel cycle, the recovery and recycling of plutonium and other actinides in reactors is recognized as the best approach for reducing the toxic waste burden on the environment. Compared to once through fuel cycles, closed fuel cycle strategies can achieve a more than 100-fold reduction of the long term radiotoxicity.

4.5. DESIGN MEASURES

Reprocessing operations generate secondary wastes. Several technical practices are commonly considered by the designers of reprocessing facilities to minimize secondary wastes. Except as noted, most of the following are applicable to both aqueous and pyro processes (however, there will always be some process loss, resulting in actinides in the waste forms):

- Enhancing the plutonium uranium redox extraction (PUREX) process (e.g. by using salt-free reagents or electro techniques to reduce the solid waste from excess sodium salt, reducing the number of cycles to simplify the process, increasing the recovery of uranium and plutonium, and reusing solvents to decrease the solvent waste);
- Integrating the separation and partitioning process;
- Reducing the solvent waste in partitioning of minor actinides (e.g. using adequate solvents composed only of carbon, hydrogen, oxygen and nitrogen to reduce solvent waste by combustion);
- Increasing the lifetime and capacity of the equipment to decrease the equipment waste;

- Applying advanced material to limit corrosion of equipment, which can decrease the equipment waste (e.g. use of complexant to reduce corrosion);
- Considering maintenance and decommissioning requirements early in the design phase to decrease the waste from operation and decommissioning;
- Recycling to decrease the resources used (e.g. water, solvent);
- Compacting or super compacting waste.

The deployment of innovative reactors that can make use of bred fissile materials (such as Pu and ^{233}U) generated from closed nuclear fuel cycle systems could reduce the amount of natural uranium needed to operate nuclear reactors. The amounts of radioactive waste from uranium mining and other stages of the nuclear fuel cycle could also be reduced proportionally. Other characteristics of waste resulting from most front end stages of conventional uranium nuclear fuel cycles would remain the same (except from enrichment).

The deployment of innovative reactors using thorium as fertile material will result in opening the scope of doubling and/or enhancing the nuclear material resources without initially generating front end cycle waste, as large stockpiles of purified thorium oxide already exist worldwide as a by-product of rare earth extraction.

Mining or milling waste is L&IL long lived waste, although it is considered low hazard waste. The large scale deployment of fast reactors could contribute to a reduction in the consumption of uranium and, hence, of the production of waste from uranium mining and milling.

4.6. WASTE STREAMS AND CHARACTERISTICS

Although it is difficult to compare wastes from innovative fuel cycles without their detailed designs, it is worthwhile to investigate potential issues in waste form or waste treatment by making a qualitative comparison of waste streams based on conceptual process flows.

As mentioned earlier, the main families of innovative reactors are sodium cooled, lead cooled, gas cooled, very high temperature, water cooled and molten salt. The characteristics of the waste from these reactors will depend on which of five chemical forms — oxide, carbide, nitride, metal or molten salt — is selected for the reactor fuel.

The technically possible combinations of reactor type and fuel type are arranged in Table 9. Possible reprocessing methods are also shown, together with the corresponding fuel fabrication methods. Those options without proper fuel fabrication methods have been screened, as shown in Tables 10–20, which reduces the total number of possibilities to 11 cases.

Spent fuel is composed of its assembly hardware, cladding, heavy metals and solvents from which only heavy metals are recycled; the other components end up as primary wastes. Solvents are divided into groups depending on their waste streams (noble gas, ^{3}H, ^{129}I, ^{14}C, noble metals (NMs), ^{99}Tc, ^{135}Cs, ^{137}Cs, ^{90}Sr and others).

Noble gas

In current fuel cycles, noble gas from spent fuel is released into the environment. Affordable techniques for capture and subsequent disposal of noble gas should be developed for use in innovative fuel cycles. Such a process is planned for the MSFR concept, which involves a continuous on-line extraction of noble gases and storage for their decay.

Tritium (^{3}H)

Tritium (^{3}H) is a product of:

- The ternary fission of uranium or plutonium in a reactor that accumulates with the increase of burnup;
- Activation products in water and helium cooled reactors;
- Normal hydrogen and deuterium activation, which is then absorbed in fuel cladding as a hydride.

In current fuel cycles, diffusion of ^{3}H from fuel is mostly trapped in the zirconium alloy cladding. A part of the ^{3}H in HLLW is vaporized into bulk tritiated water during the fission product concentration process. A public concern is posed by ^{3}H because it is a bioactive element that may interfere with biological cycles; therefore, its release or storage must be considered. Tritiated coolant and metal need to be dealt with during decommissioning. It is reasonable to store the separated tritium for decay (its half-life is 12.6 years) if it can be concentrated into a small volume stream.

Iodine-129 (^{129}I)

In current fuel cycles, iodine is absorbed on silver loaded absorbents and must be conditioned. There is room for improvement in waste form to decrease its release rate, as the bioactivity, long half-life, and mobility of ^{129}I is rather high. In innovative fuel cycles based on some pyro reprocessing, iodine may mix with chlorine and molten salt or fluoride salts from MSRs. Development work on separation and efficient immobilization of iodine needs to be implemented; the ceramic form might be a good choice. (Note that the option of sea disposal of ^{129}I is the management strategy for both Sellafield and La Hague, and although a technically sound approach, it might be a politically problematic one.)

Carbon-14 (^{14}C)

In current fuel cycles, small amounts of ^{14}C are discharged into the environment as gas or liquid. With the use of nitride fuel, it is important to enrich nitride in ^{15}N to avoid generating a large amount of ^{14}C from ^{14}N being 99.6% of natural abundance. To date, some studies have examined the immobilization of ^{14}C in a carbonate waste form or in ceramic, but further research is needed as these discharges may raise public concern. In addition, activation of ^{14}N and ^{13}C in a gas reactor generates ^{14}C. The ^{14}C is also produced from ^{17}O, which is of concern for oxide fuels.

Technetium-99 (^{99}Tc)

In current fuel cycles, the fission product ^{99}Tc may become part of airborne, liquid or solid wastes; after spent fuel reprocessing; it is vitrified in glass. Due to the long half-life and high mobility of TcO_4 in oxidizing environments it is a problematic nuclide. Innovative nuclear fuel cycles should solidify ^{99}Tc into a suitable waste form or transmute it.

Noble metals

Noble metals are generally recovered in fine epsilon metal particles from the feed clarification stage of reprocessing. To avoid interference with the vitrification process, these particles can be immobilized in alloy form or incorporated into other waste forms.

Caesium/strontium (Cs/Sr)

Caesium and strontium are the major heat contributors to the glass waste forms for geological repositories. In innovative nuclear fuel cycles, separating caesium/strontium for solidification and decay storage may be able to decrease the footprint of geological disposal. Alternatively, HLW forms with higher heat tolerance can be developed.

Secondary wastes such as solvent, chemicals and contaminated materials are also listed in Tables 10–20 for each of the screened fuel cycle options. Corresponding waste streams and waste forms are described, together with the status of the technology to make their waste forms. They are categorized into the following five stages of development:

— *Mature:* The technology is commercially deployed with similar waste streams.
— *Industrial:* The technology exists at an industrial scale and is considered a likely candidate.
— *Pilot:* The technology exists at a pilot scale and is considered a good candidate for industrialization.

— *Laboratory:* The technology is under development at a laboratory scale.
— *Conceptual:* The process depends on theoretical consideration or contains some speculation.

The potential issues of each waste stream are also described in the tables where important R&D needs are identified by hatching in the cell. Using this kind of systematic approach, fuel cycle options can be assessed from the waste stream viewpoint.

The following sections discuss the two methods of reprocessing spent fuel — aqueous and pyro — from the innovative reactors listed in Table 9.

4.7. FUEL CYCLE BASED ON AQUEOUS REPROCESSING

Aqueous reprocessing refers to recovering the heavy metals from the aqueous solution of spent fuel with chemical methods — usually extraction. The PUREX process is a well known aqueous process for industrial scale spent fuel reprocessing (see Annex 1 for details). Generally, the spent fuel of innovative reactors will contain a large amount of plutonium, so the extracted feed solution might be diluted.

All aqueous processes consist of four main steps: head end (including dissolution), off-gas cleanup, solvent extraction and vitrification of HLLW. The processes in Cases 1 through 7 are almost the same, except for some differences in the head end step, although the spent fuel in innovative reactors differs significantly. A general flow of the aqueous processes, together with the waste streams, is shown in Fig. 6.

The major difference between the reprocessing of oxide fuel, carbide fuel and nitride fuel exists in the head end. Oxide fuel is dissolved by nitric acid then clarified, which produces feeding solution for subsequent extraction, insoluble residue from the clarification, and nitrogen oxide off-gas. Carbide and nitride fuels may require an additional operation before PUREX, such as conversion into oxide form by combustion or oxidation. Off-gas from the de-cladding and dissolution processes is contaminated with ^{3}H, ^{14}C, ^{15}N and ^{129}I, and released of noble gas. The amount of released ^{14}C or ^{15}N depends on whether the fuel is carbide or nitride.

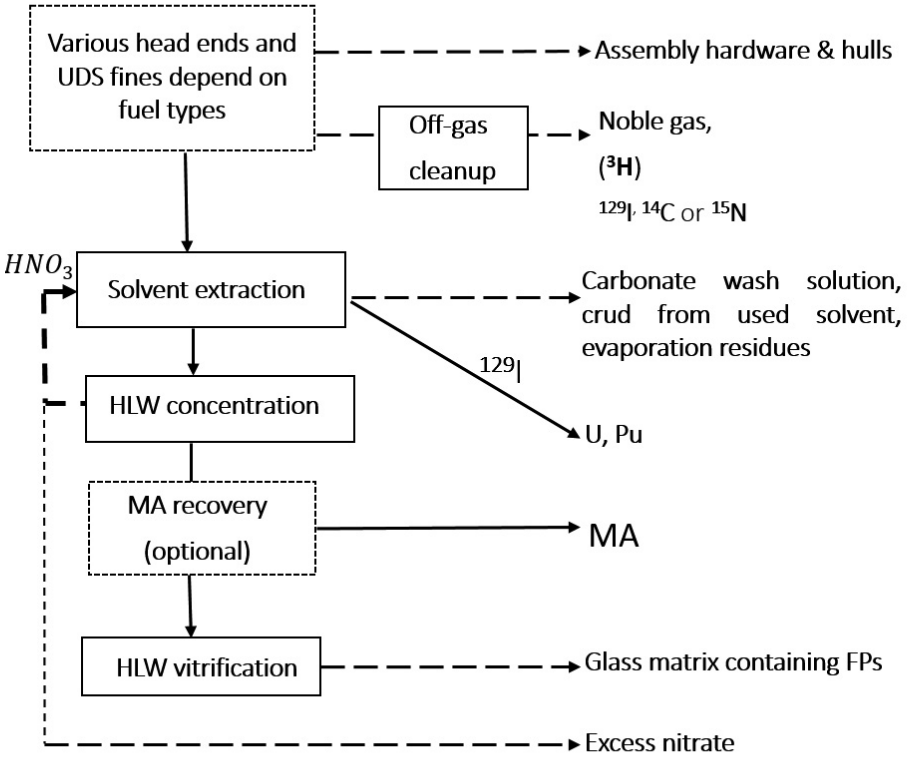

FIG. 6. The flowsheet and waste stream of an aqueous process. FP — fission product; HLW — high level waste; MA — minor actinide; UDS — undissolved solid.

Off-gas is usually treated in the off-gas cleaning system, which generates waste in a form of spent iodine-bearing silver substrates, caustic scrub solutions and/or spent molecular sieves, as well as some ^{14}C or ^{15}N.

The quantity of purified and released krypton is approximately the same in all cases. Solvent extraction, based on the PUREX process, routinely generates waste streams, including HLLW, carbonate wash solution, crud from used solvents, and evaporator condensate. The treatment of these waste streams is well known; two options for treatment of HLLW could be used. Part of the recovered nitric acid returns to the dissolver for the dissolution of oxide fuel, whereas excess acid is released or finally conditioned.

The vitrification process is the primary option for handling HLLW from aqueous reprocessing, which consists of fission products and minor actinides. It is the only proven technology for such waste, with extensive data on its impact on the safety of geological disposal. Vitrification of the HLLW forms a glass matrix which immobilizes the fission products and minor actinides.

Another option under R&D is to partition out the minor actinides before vitrification, as they may introduce long term risk to the geological repository. Partitioning of minor actinides for recycling or transmutation would result in a significant decrease in transuranic (TRU) inventory within the waste. However, it is not easy to make an adequate comparison among special processes developed for the partitioning of minor actinide from the stream.

In the following sections, waste streams from the reprocessing of used fuel from each of the innovative reactors is addressed.

4.7.1. Waste from the aqueous fuel cycle of liquid metal cooled reactors

Case 1: Oxide fuel

The waste streams and characteristics are described in Table 10 (Case 1). Since this option is an application of the current fuel cycle technology for LWR spent fuel to the fast reactor (FR) spent fuel, the potential issues related to waste are:

- Higher radioactivity or amount of the primary waste (structural materials and HLLW);
- Hydrogen gas generation if the cement process is used for conditioning assembly hardware and hulls (these metallic wastes should be compacted or melted);
- Impacts on vitrification with the incorporation of higher concentrations of noble metal into the glass or an alternative approach to manage undissolved solids (e.g. cold crucible technology); specifications must be extended for the content of alpha activity in the glass.

A typical reprocessing process flow with the waste streams is shown in Fig. 7. In the disassembly process, the wrapper tube must be removed before chopping the fuel pins. The amount of assembly hardware which becomes solid waste is larger than for LWR spent fuel. The noble gas stream ^{14}C, and ^{129}I are released as the off-gas from the fuel pin chopping process and dissolution process. In FR spent fuel, all tritium from the oxide and cladding is diffused to the sodium coolant of the reactor. This is a significant difference from LWR fuel, where tritium is distributed between the oxide fuel and the Zircaloy cladding.

The cladding hulls are rinsed and compacted into solid waste. The insoluble residues are separated in the clarification process and combined with HLLW. The solvents can be recycled after being washed by Na_2CO_3 or salt-free chemicals to eliminate degradation products such as dibutyl phthalate (DBP) and monobutyl phthalate (MBP) which become solid wastes that should be retained, as after-alkaline hydrolysis of spent organic generated waste can be fixed in cement. Since vitrification is the only proven technology for the waste, and extensive data exist to evaluate the safety of its geological disposal, it should be the first candidate for the conditioning of HLLW from aqueous reprocessing. The process for recovery of minor actinides is under development, but the goal of the research is to develop a molecule resistant to hydrolysis and radiolysis that can be recycled after regeneration (like tri-n-butyl phosphate (TBP)). Research is focused primarily on molecules containing carbon, hydrogen, oxygen, sulphur and nitrogen (i.e. those that are phosphate free).

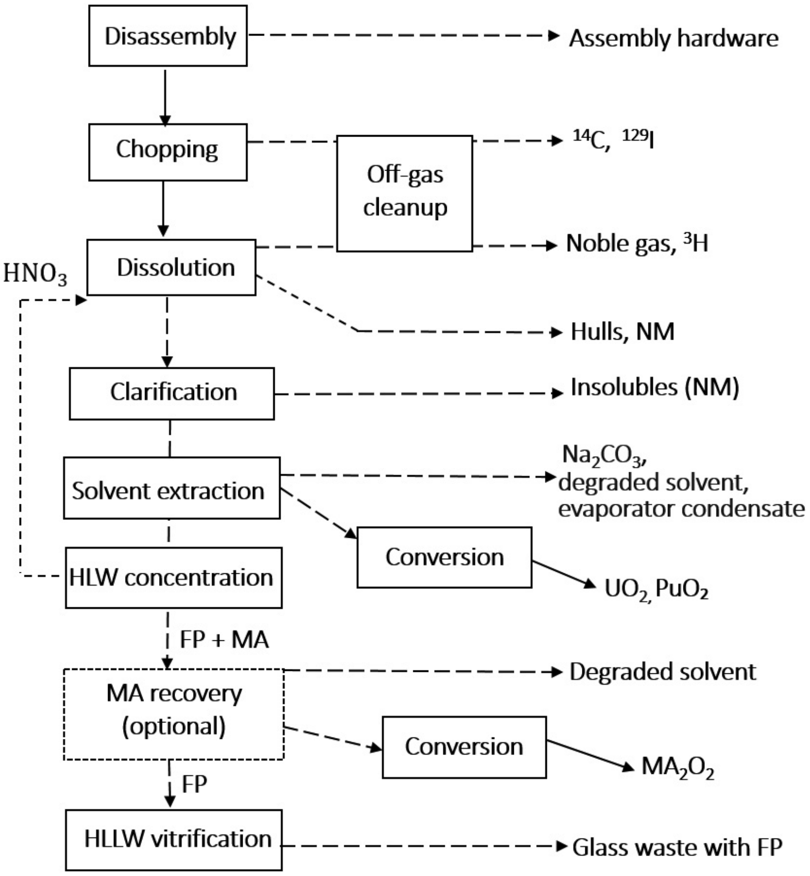

FIG. 7. Process flow and primary waste stream for Case 1 (FR/O/Aq). FP — fission product; HLLW — high level liquid waste; HLW — high level waste; MA — minor actinide; NM — noble metal; UDS — undissolved solid.

Case 2: Carbide fuel

The waste streams and characteristics are described in Table 11 (Case 2). In addition to the potential issues with oxide fuel (Case 1), a potential issue for carbide fuel relates to wastes from the off-gas treatment, if combustion is used before dissolution.

A typical reprocessing process flow is shown in Fig. 8, together with the waste streams. The PUREX process can be applied to carbide fuels after their conversion to oxide form. The process is composed of four steps: head end, off-gas cleanup, solvent extraction and HLLW treatment. The actinide carbide compounds are highly susceptible to oxidation and hydrolysis and are pyrophoric in powder form. The entire fabrication and head end processes therefore need to be carried out inside hot cells in an inert cover gas (N_2, Ar, He, etc.) atmosphere containing minimal amounts of oxygen and moisture (<20 ppm each).

Advanced fuels can be converted to oxide form through a wet chemical process, but this involves the production of complex organic molecules in a dissolution solution, which interferes with the plutonium during the extraction process and results in leakage of plutonium in HLLW and the solvent after the back-extraction of plutonium. Voloxidation may be used to directly burn the carbides to oxides. Researchers from India have proposed electro-oxidative dissolution to destroy these organic compounds, as the gaseous oxidation process does not have this disadvantage [30, 31]. For easy reprocessing, the oxidized fuel must be exclusively close to stoichiometric (uranium, plutonium) O_2, containing 75% uranium and 25% plutonium. The subsequent combustion of the fuel leads to the generation of a large amount of carbon dioxide off-gas contaminated with ^{14}C, ^{129}I and semi-volatile radionuclides and noble gas. Like FR oxide fuel, carbide spent fuel (like nitride fuel and all FR ceramic fuel with stainless steel cladding) does not contain tritium because it is completely diffused from the fuel to the sodium coolant of the reactor.

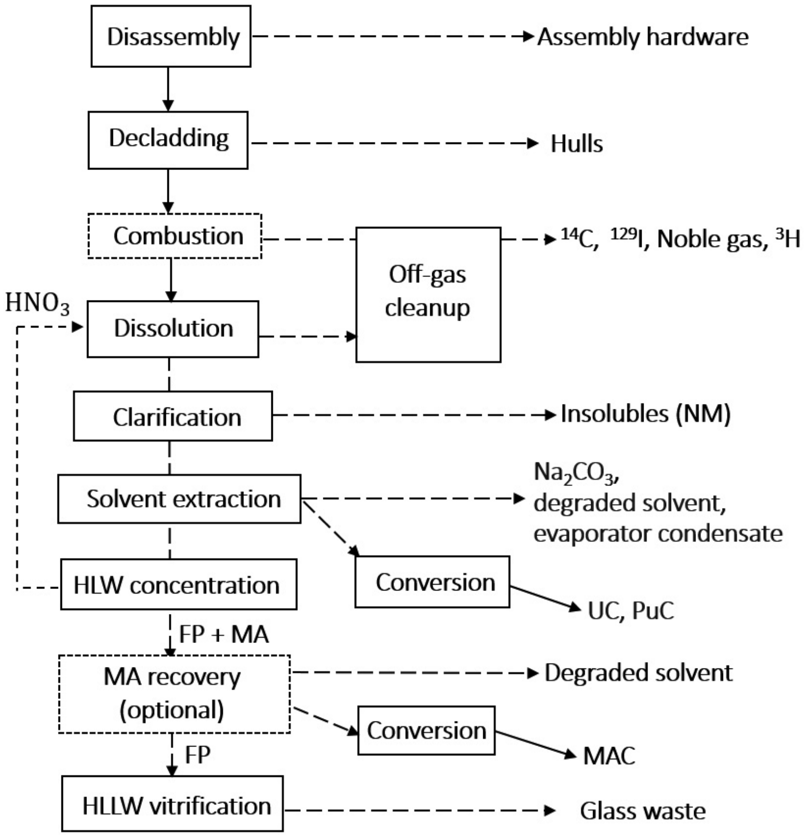

FIG. 8. Process flow and primary waste stream for Case 2 (FR/C/Aq). FP — fission product; HLLW — high level liquid waste; HLW — high level waste; MA — minor actinide; NM — noble metal; UDS — undissolved solid.

In the case of combustion, off-gas needs careful treatment in the off-gas cleaning system because it generates waste in the form of iodine-bearing silver substrates, caustic scrub solutions and/or spent molecular sieves and ^{85}Kr. The combustion of C in CO$_2$ leads to a large dilution of ^{14}C in the gas. The production of large amounts of CO$_2$ contaminated with ^{14}C increases the amount of carbonate waste as a result of scrubbing. The other waste streams are the same as those mentioned in Case 1. Application of the PUREX process generates the same waste streams, including HLLW, carbonate wash solution, crud from used solvents and evaporator condensate. The treatment of these waste streams is well known.

Case 3: Nitride fuel

The waste streams and characteristics are described in Table 12 (Case 3). The potential issues related to waste other than those mentioned in Case 1 are:

- High concentrations of ^{15}N: its recycling will be necessary to avoid the generation of ^{14}C from ^{14}N.
- Wastes from the oxidation step before dissolution to recover ^{15}N are not well known.

A typical reprocessing process flow is shown in Fig. 9, together with the waste streams. Like carbide, nitride compounds are highly susceptible to oxidation and hydrolysis and are pyrophoric in powder form. The main difference between the reprocessing of nitride fuel and oxide fuel is the need to convert nitride into oxide. After conversion to oxide form, the PUREX process can be applied to the reprocessing of nitride fuel. This generates the same PUREX waste as in Case 1. However, a very large amount of ^{14}C can be produced by the ^{14}N(n,p)^{14}C reaction in the reactor. To avoid this problem, isotopic concentration of ^{15}N of 99.9% is proposed, but it should be recycled since the cost is high. In this case, released ^{15}N in the oxidation step must be recovered from the off-gas and recycled in the conversion step of actinides in nitride form.

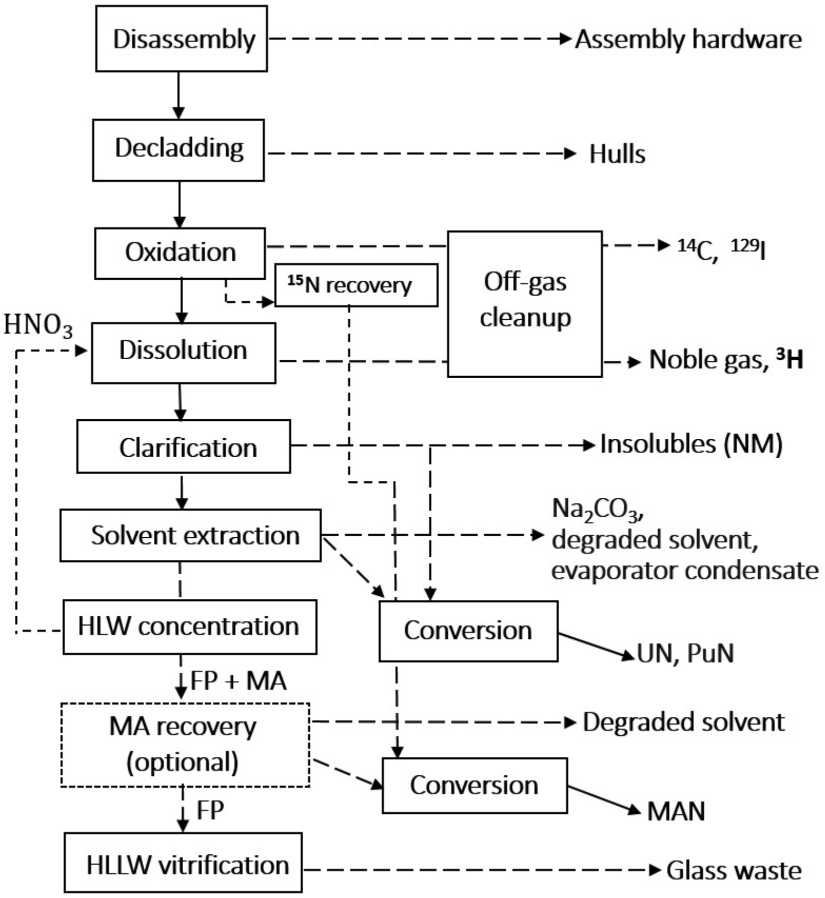

FIG. 9. Process flow and primary waste stream for Case 3 (FR/N/Aq). FP — fission product; HLLW — high level liquid waste; HLW — high level waste; MA — minor actinide; NM — noble metal; UDS — undissolved solid.

If the recovery of ^{15}N is not realized before the nitric dissolution of spent fuel, NO_2 is produced with ^{15}N, ^{129}I, semi-volatile radionuclides and noble gas. Direct dissolution of nitride fuel into HNO_3 is possible, but it can be difficult to recover and purify the ^{15}N during these steps.

4.7.2. Waste from the aqueous fuel cycle of VHTRs

Case 4: Oxide or carbide fuel

Future gas cooled reactors will almost certainly utilize coated particle fuel (Table 13, Case 4). This fuel consists of oxide or carbide; uranium, plutonium, or U–Th compounds in the form of microspheres from 50 to at least 500 µm in diameter, with successive coatings of porous carbon, pyrolytic graphite, silicon or zirconium carbide; and a final outer coating of pyrolytic graphite. The overall particle diameter is on the order of 800 µm to 1 mm. With the unique structure of the particle fuel, difficulties are related to the accessibility of the uranium kernels, which are coated in carbon and SiC layers and dispersed in a large volume of graphite. A typical reprocessing process flow is shown in Fig. 10, together with the waste streams.

The VHTR has been proposed to operate in an open fuel cycle, but if the fuel is to be reprocessed to reduce radiotoxicity, these coating layers must be removed in order to provide reagent access to the fuel material. The mechanical extraction of compacts from the spent fuel blocks appears to be promising, followed by removal of the graphite from the compacts using pulsed currents that can free the particles. Subsequently, the carbon and silicon carbide layers can be removed by high temperature oxidation or by carbo-chlorination to access the kernels. Or the kernels can be exposed by jet milling to break the multi-layered coatings. After the kernels have been separated from the different coating layers, the oxide or carbide kernels can be treated by aqueous processes as in Cases 1 or 2. The recovered actinides can be used in fast reactors.

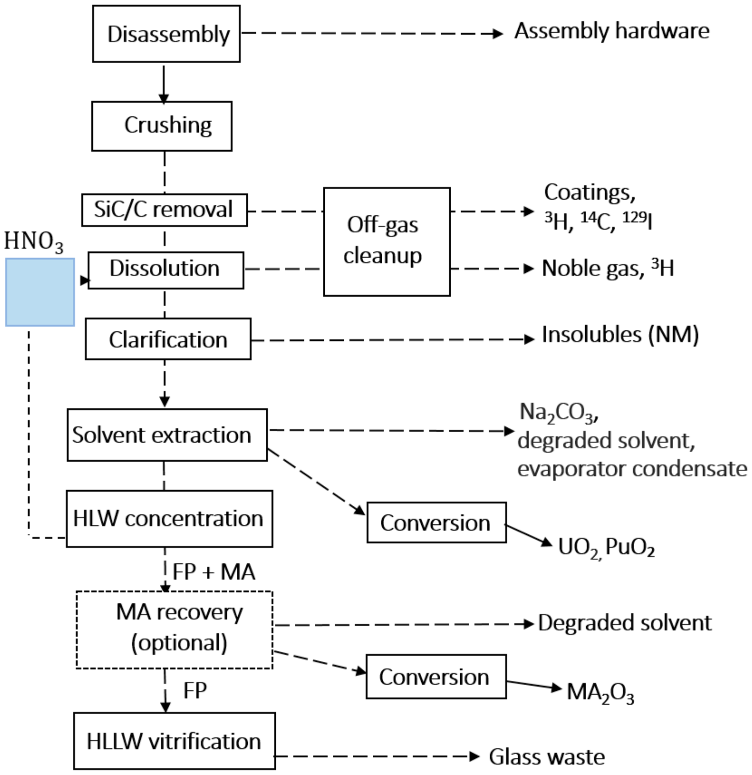

FIG. 10. Process flow and primary waste stream for Case 4 (VHTR/O/Aq). FP — fission product; HLLW — high level liquid waste; HLW — high level waste; MA — minor actinide; NM — noble metal; UDS — undissolved solid.

In the case of pebble bed fuel, feasibility tests to fragment fuel spheres and the coated fuel particles by using welding techniques showed promise. After fragmentation of the fuel particles, the fragmentation product can be directly fed into classical aqueous reprocessing in which the fuel dissolves in nitric acid. However, much further research and demonstration are needed.

The problematic waste of VHTR fuel is associated primarily with the activated graphite waste generated by the disassembly of the prismatic block and the crushing of fuel compacts. This produces a large amount of waste. There is also a significant increase in ^{14}C from the activation of ^{13}C and ^{14}N in the waste. Removal of the carbon and SiC coatings will result in large amounts of carbon waste or an additional fractured coating waste stream.

4.7.3. Waste from the aqueous fuel cycle of GCFRs

GCFR fuel will be constituted from carbide or nitride pellets clad with SiC ceramic tubing or plating. A mechanical treatment is necessary to access the carbide or nitride fuel before the nitric acid is dissolved. The major wastes of the GCFR fuel cycle are expected to be the SiC and the waste streams resulting from the separation and purification of actinide from spent carbide or nitride fuel by the PUREX process. Waste streams and characteristics are described only for carbide fuel, but the potential issues for nitride fuel are similar to those mentioned in Case 4.

Case 5: Carbide fuel

The waste streams and characteristics are described in Table 14 (Case 5). The potential issues related to waste are similar to those mentioned in Case 2.

A typical reprocessing process flow is shown in Fig. 11, together with the waste streams. The process is composed of four steps: head end, solvent extraction, off-gas cleanup and HLW treatment. In the head end, the mechanical treatment and combustion of carbide fuel and consequent dissolution lead to the generation of solid waste, SiC structure material, insoluble residue and a large volume of off-gas (CO_2), ^{129}I, semi-volatile radionuclides and noble gas. Therefore, the off-gas needs careful treatment in the off-gas cleaning system. It produces waste in

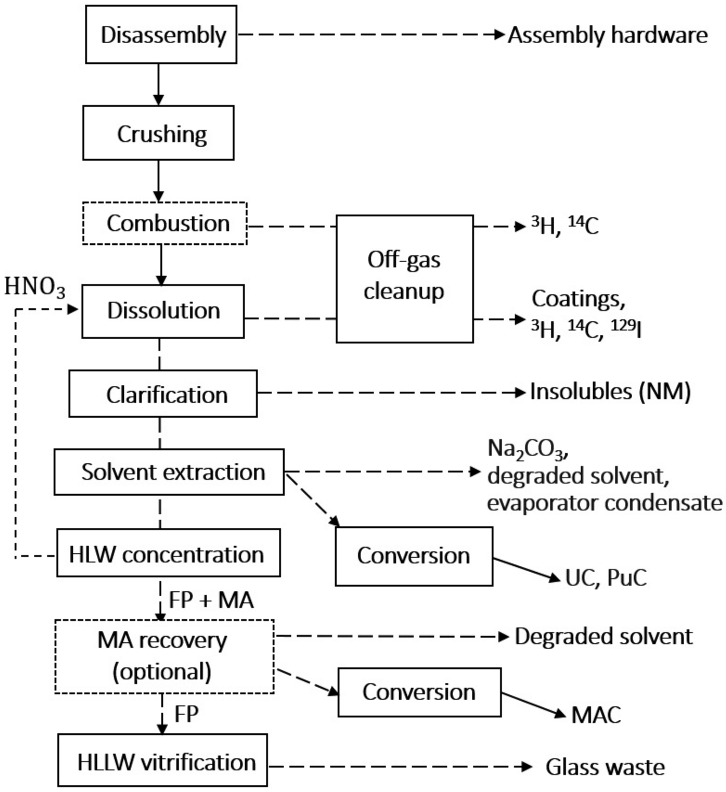

FIG. 11. Process flow and primary waste stream for Case 5 (GCFR/C/Aq). FP — fission product; HLLW — high level liquid waste; HLW — high level waste; MA — minor actinide; NM — noble metal; UDS — undissolved solid.

the form of spent iodine-bearing silver substrates, caustic scrub solutions and/or spent molecular sieves, ^{85}Kr and $^{14}CO_2$ stream. During the extraction system, the same wastes are produced as during routine PUREX. This includes HLLW, carbonate wash solution, crud from used solvents, and evaporator condensate. The other waste streams are almost the same as in Case 1.

Case 6: Nitride fuel

The waste streams and characteristics are described in Table 15 (Case 6). The potential issues related to waste are similar to those mentioned in Case 3.

A typical reprocessing process flow is shown in Fig. 12, together with the waste streams. The process is composed of four steps: head end, solvent extraction, off-gas cleanup and HLW treatment. In the head end, the mechanical treatment and oxidation of the nitrite fuel and consequent dissolution lead to the generation of solid waste, SiC structure and insoluble residue, and a large volume of off-gas contaminated with ^{14}C, ^{129}I, semi-volatile radionuclides and noble gas. The off-gas needs careful treatment in the off-gas cleaning system since it produces waste in the form of spent iodine-bearing silver substrates, caustic scrub solutions, spent molecular sieves and a $^{14}CO_2$ stream. During the extraction process, the same wastes are produced as during routine PUREX. These wastes include HLLW, carbonate wash solution, crud from used solvents, and evaporator condensate. The other waste streams are almost the same as in Case 1. However, a large amount of ^{14}C can be produced by the ^{14}N(n,p)^{14}C reaction in the reactor. To avoid this, isotopic concentration of ^{15}N by 99.9% is proposed; due to high costs, it should be recycled. In this case, released ^{15}N in the oxidation step must be recovered from the off-gas and recycled in the conversion step of actinides in nitride form.

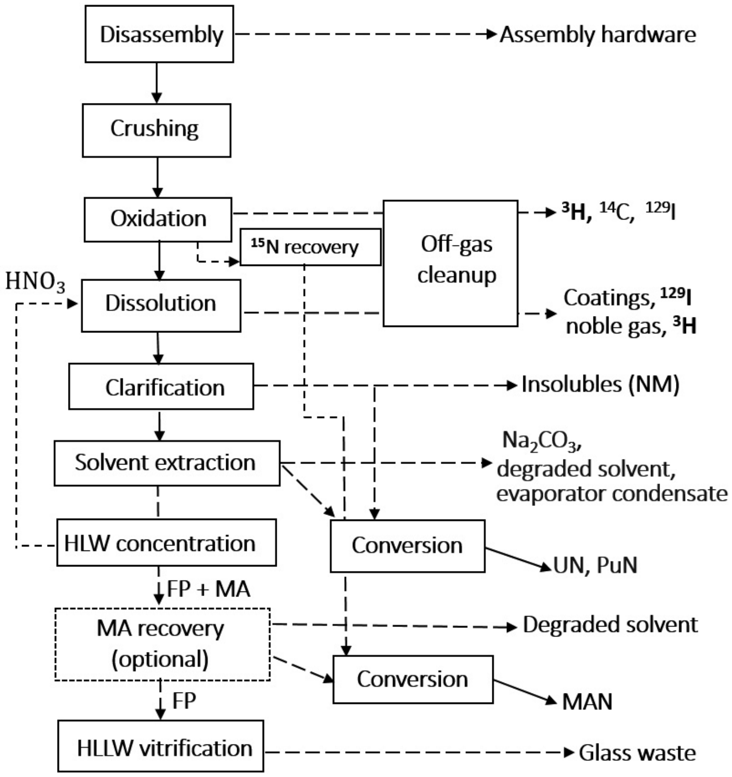

FIG. 12. Process flow and primary waste stream for Case 6 (GCFR/N/Aq). FP — fission product; HLLW — high level liquid waste; HLW — high level waste; MA — minor actinide; NM — noble metal; UDS — undissolved solid.

4.7.4. Waste from the aqueous fuel cycle of AHWRs

Case 7: Oxide fuel

The waste streams and characteristics of oxide fuel are described in Table 16 (Case 7). A typical reprocessing process flow is shown in Fig. 13, together with the waste streams.

The process flow and its waste streams are identical with Fig. 6 for Case 1 (oxide fuel with aqueous reprocessing), except for the different structural material and fuel (U–Th oxide, Pu–Th oxide).

Most of the constraints associated with the thorium fuel cycle are found in spent fuel reprocessing and the subsequent handling of the separated ^{233}U and Th products. The thorium–uranium extraction (THOREX) process for the recovery and recycling of ^{233}U and Th has several special requirements. In contrast to UO_2, crystalline ThO_2 and unirradiated PuO_2 are difficult to dissolve in nitric acid without the addition of appropriate amounts of fluoride. However, the use of fluoride is undesirable because of its corrosive nature and incompatibility with common materials in reprocessing equipment.

The (n,2n) reactions encountered during the irradiation of thorium lead to the formation of long lived ^{231}Pa and relatively short lived (68.9 years) ^{232}U, with its hard beta gamma emitting decay products. Thus, the ^{233}U produced in the reactor is contaminated with ^{232}U. The level of contamination depends on the burnup and on the neutron spectrum encountered in the reactor. The radiological hazard in handling the reprocessed ^{233}U arises mainly from the alpha and gamma activities associated with the ^{232}U isotope formed during irradiation and its short lived decay products. These hazards lead to the necessity of using remote handling and fabrication techniques.

4.8. FUEL CYCLE BASED ON PYRO PROCESSING

Another option for reprocessing the spent fuel from innovative reactors is pyro processing, which refers to recovering the uranium and TUE from spent fuel in a molten salt medium under high temperature by means

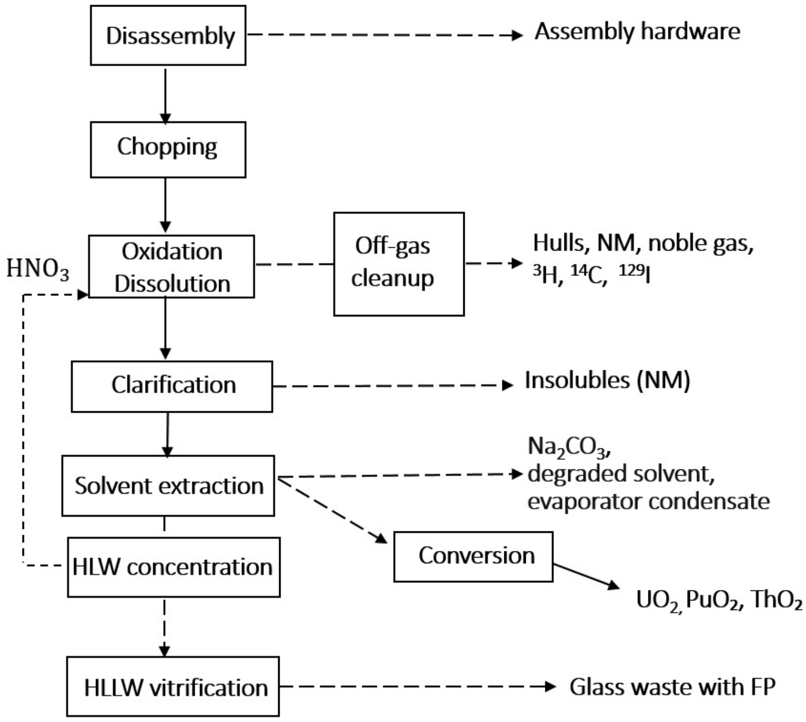

FIG. 13. Process flow and primary waste stream for Case 7 (AHWR/O/Aq). FP — fission product; HLLW — high level liquid waste; HLW — high level waste; MA — minor actinide; NM — noble metal; UDS — undissolved solid.

of electrochemical methods or molten salt extraction. This process is developed for metallic fuel, oxide fuel and nitride fuel. Only a conceptual design is proposed for fluoride fuels (Section 4.8.2: Case 11).

In the electro-refining process, metallic fuel pins are chopped and the fuel pin segments are placed in a stainless steel mesh basket, which becomes the anode of an electro-refining cell using an LiCl–KCl electrolyte. In the case of oxide fuels, UO_2 and MOX are dissolved into molten salt either by chlorine gas or oxide reduction followed by anodic dissolution. Uranium is electro-transported from the salt to the steel cathode. The TRUs will not deposit on the steel cathode unless the concentration is about 100 times higher than that of uranium, as TRU Cl_3 is more stable than UCl_3 in the salt. The TRU can be recovered by a different cathode, one in which the TRUs deposit as inter-metallic compounds with cadmium in a crucible containing liquid cadmium that is suspended in salt. Deposition of the TRUs is accompanied by a certain amount of uranium, depending upon its concentration in the salt, typically being a mixture of 70% TRUs, 25% uranium and 5% lanthanide fission products.

The TRUs are left in the spent salt, which will be periodically removed to produce a composite glass–ceramic waste after recovering residual TRUs, if needed and possible. The noble metal fission products that remain in the anode basket are combined with the cladding hulls and melted together to form a metallic waste for disposal.

Nitride fuels can also be anodically dissolved into molten salt, but the deposit metallic uranium or co-deposit uranium, plutonium and minor actinides in a cadmium cathode must be converted to nitride. A generalized flowsheet of the pyro processes is shown in Fig. 14, together with the waste streams.

4.8.1. Waste from the pyro fuel cycle of liquid metal cooled reactors

Case 8: Oxide fuel

The waste streams and characteristics are described in Table 17 (Case 8). The potential issues related to waste, other than those described for Case 1 (except vitrification), are:

- Recycling of the chlorine off-gas, which contains ^{129}I;
- High temperature alloying of the noble metal;

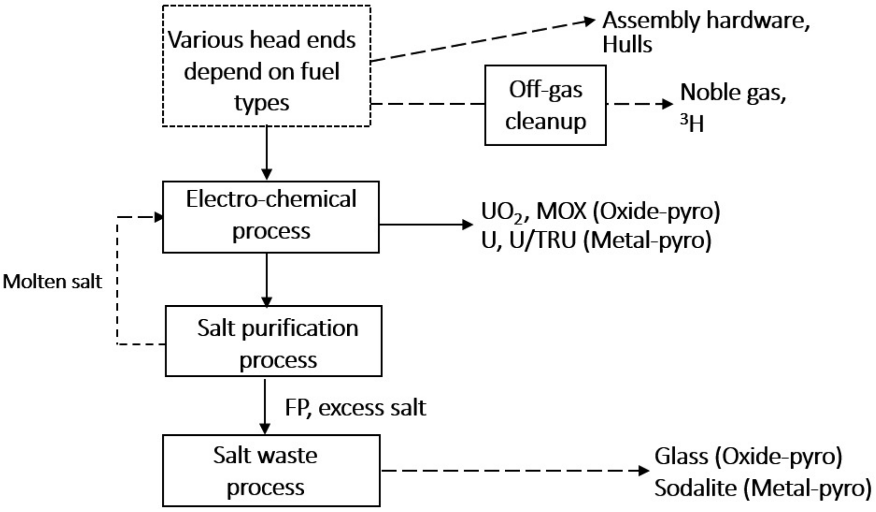

FIG. 14. Generalized flowsheet and waste stream of pyro processes. FP — fission product; MOX — mixed oxide; TRU — transuranic.

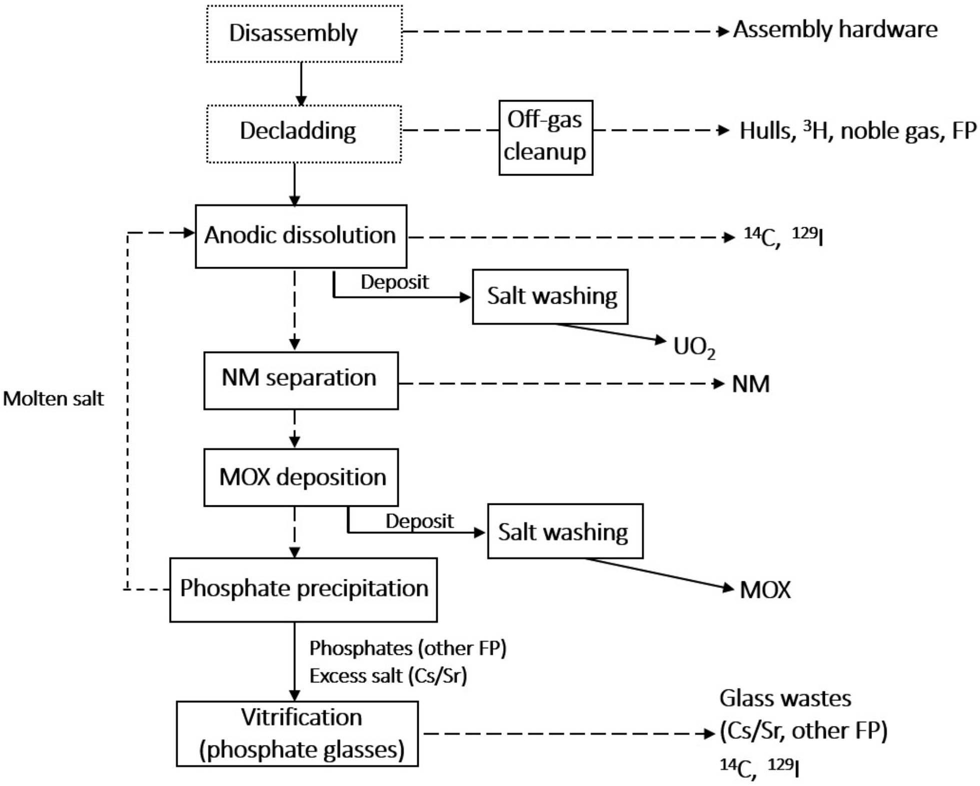

FIG. 15. Process flow and primary waste stream for Case 8 (FR/O/Py). FP — fission product; MOX — mixed oxide; NM — noble metal.

— Incomplete oxide reduction/dissolution results in significant UOX in the metal waste;
— Cadmium waste management;
— Immobilization of the salt waste.

A typical reprocessing process flow is shown in Fig. 15, together with the waste streams. The cladding should be separated to avoid its chlorination. The separated hulls are rinsed with molten salt to recover heavy metals adhering to them. Noble gas is released from the de-cladding or dissolution process, which is released as gas waste from the chlorine gas after treatment (cryogenic recovery process). The behaviour of ^{129}I in the dissolution process is not well known. Since iodine and chlorine belong to the halogen group, they could coexist in the molten salt or chlorine gas. There may be ^{14}C released in a noble gas stream. The noble metal fission products are deposited onto the solid cathode made of noble metal. The deposited noble metals are alloyed by induction heating. Dissolved uranium exists as oxide ions and is deposited as UO_2 on to the solid cathode. Then plutonium is oxidized to oxide ions and deposited as MOX onto the solid cathode. As oxide ions of minor actinides are not stable, a recovery process for minor actinides has not yet been developed.

Case 9: Nitride fuel

The waste streams and characteristics are described in Table 18 (Case 9). The potential issues related to waste, other than those described for Case 1 (except vitrification), are:

— High concentrations of ^{15}N; its recycling will be necessary to avoid the generation of ^{14}C from ^{14}N;
— Unknown behaviour of ^{129}I and ^{14}C;
— Cadmium waste management;
— High temperature alloying of the noble metals;
— Reduction of the salt waste volume.

A typical reprocessing process flow is shown in Fig. 16, together with the waste streams. The process flow is very similar to that for metallic fuels (Case 10). As there is no bonding of sodium, the chopped fuels are loaded in baskets to be immersed and electrochemically dissolved (anodic dissolution) in the molten salt, leaving the hulls and the noble metals, including ^{99}Tc, in the anode basket. Only uranium metal is deposited on the solid cathode. Then uranium, plutonium and minor actinides are co-deposited into the liquid cadmium cathode, leaving the others in the salt. The amount of ^{14}C can be very large due to its production by a $^{14}N(n,p)^{14}C$ reaction in the reactor. An isotopic concentration of ^{15}N up to 99.9% is proposed to be used as nitride fuel, but because of high costs, the ^{15}N should be separated from the off-gas and recycled in the conversion step.

The rest of the process and waste streams are yet to be studied, but are expected to be very similar to Case 3.

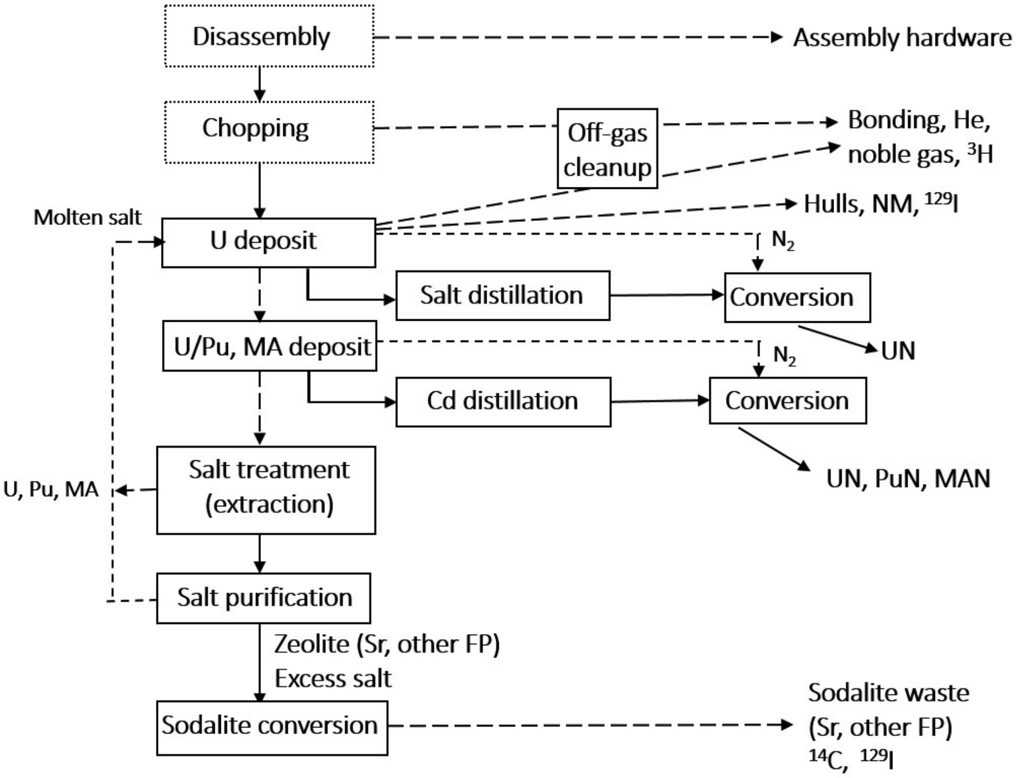

FIG. 16. Process flow and primary waste stream for Case 9 (FR/N/Py). FP — fission product; MA — minor actinide; NM — noble metal.

Case 10: Metallic fuel

The waste streams' characteristics are described in Table 19 (Case 10). The potential issues related to waste, other than those described for Case 1 (except vitrification), are:

— Unknown behaviour of ^{3}H, ^{129}I and ^{14}C;
— High temperature alloying of the noble metals (the alloying of metal waste has been demonstrated at full scale with fully radioactive waste);
— Cadmium waste management;
— Reduction of the salt waste volume.

A typical reprocessing process flow is shown in Fig. 17, together with the waste streams. In order to decrease the amount of waste salt, the bonding sodium in the chopped fuel pins is removed by distilling it with caesium. Then the treated chopped pins are loaded into baskets to be immersed and electrochemically dissolved (anodic dissolution) in the molten salt. Only uranium metal is deposited on the solid cathode. Then uranium, plutonium and minor actinides are co-deposited into the liquid cadmium cathode, leaving the others in the salt. The bonded Na will contain ^{129}I, and so will ultimately need to be put into HLW form. The alternative is to leave it with the fuel pieces in the electro-refiner (ER).

The hulls are rinsed with molten salt to recover heavy metals adhering to it. Noble gas and ^{3}H are released as waste from the distillation or dissolution process. The behaviour of ^{129}I in the dissolution process is not well known. Since iodine belongs to the halogen group, it could stay in the molten salt as iodides. Noble metals will not be dissolved by the anodic dissolution and will stay in the basket or fall into the bottom of the electro-refiner or the cadmium pool. Fine particles of ^{14}C (carbide or carbon) may be released into the molten salt, which will be disposed of with the excess salt. In order to keep the concentration of others in the molten salt at a certain level, a part of the spent salt will be removed and purified before being absorbed onto a zeolite column. Uranium, plutonium and minor actinides in the salt will be extracted prior to the purification and returned into the molten salt

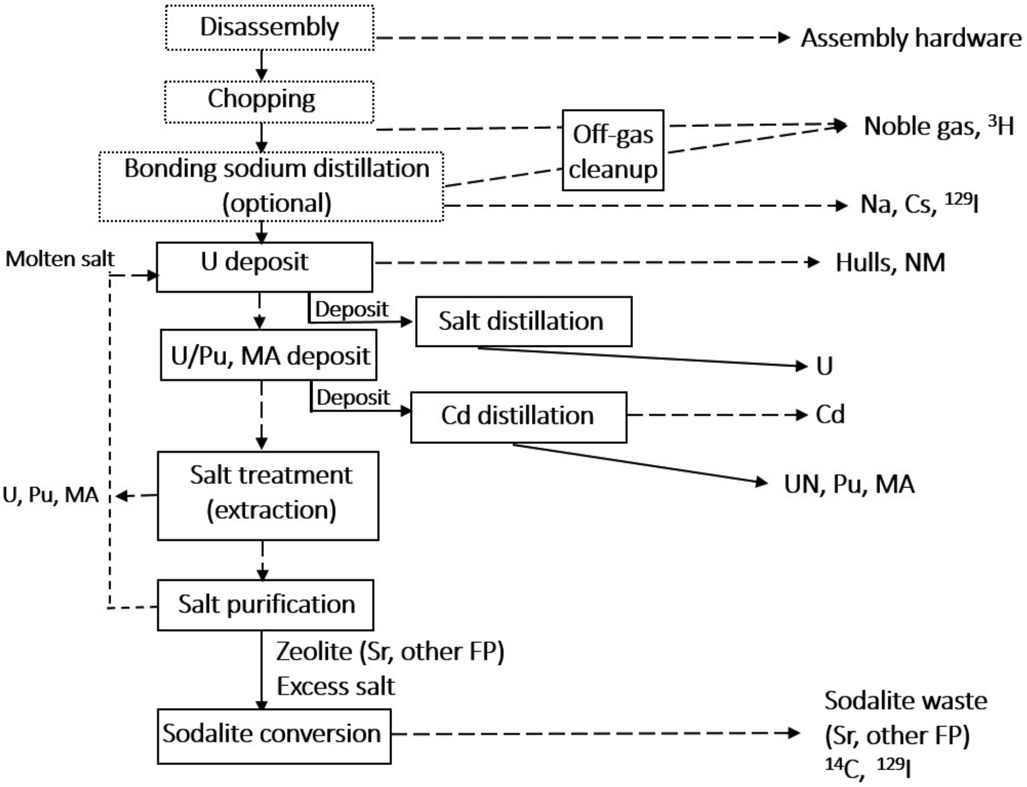

FIG. 17. Process flow and primary waste stream for Case 10 (FR/M/Py). FP — fission product; MA — minor actinide; NM — noble metal.

after the purification. The removed salt in the zeolite will be thermally converted to sodalite with the excess salt, then removed by bonding with sodium and caesium after oxidation, and crushed in moulds if used for fuel casting or as a borosilicate glass additive. Here, ^{14}C is assumed to be produced from a ^{14}N impurity in the cover gas of a fuel fabrication furnace.

4.8.2. Waste from the pyro fuel cycle of molten salt fast reactors (MSFRs)

Case 11: Molten salt fuel with pyro processing

Reprocessing U–Th fuel in an MSR is a process that has not been well developed (Table 20, Case 11). Previous demonstrations and studies have been done for thermal neutron reactors (aircraft reactor experiment (ARE), molten salt reactor experiment (MSRE), molten salt breeder reactor (MSBR), in Oak Ridge, Tennessee, USA) and with salt containing beryllium or zirconium fluoride. The MSFR reprocessing scheme currently proposed is based on data accumulated from 1952 to 1975, but differs from the one proposed for the MSBR by the quantity and some of the species to be extracted.

In the MSFR case (a short description of the reactor concept is given in Annex II), molten fuel reprocessing combines an on-line continuous gaseous extraction system and an on/off-line lanthanide extraction process (Fig. 18). The continuous extraction system will also remove most of the NM particles present in the fuel flow. The presented scheme is a general outline for the processing stage (about 10 to 40 l/day — an absolute periodicity is not required). Without further studies, it is difficult to provide detailed descriptions of each stage, but the main difficulties might lie in the tritium confinement and the complications for reprocessing due to the presence of zirconium. Elements such as protractinium, which created problems for MSBR salt processing, do not disturb MSFR reprocessing.

Fundamental data for reprocessing still need to be acquired, but numerous stages cover subjects common to classical pyrochemical solid fuel reprocessing studies, especially the separation of actinides and lanthanides.

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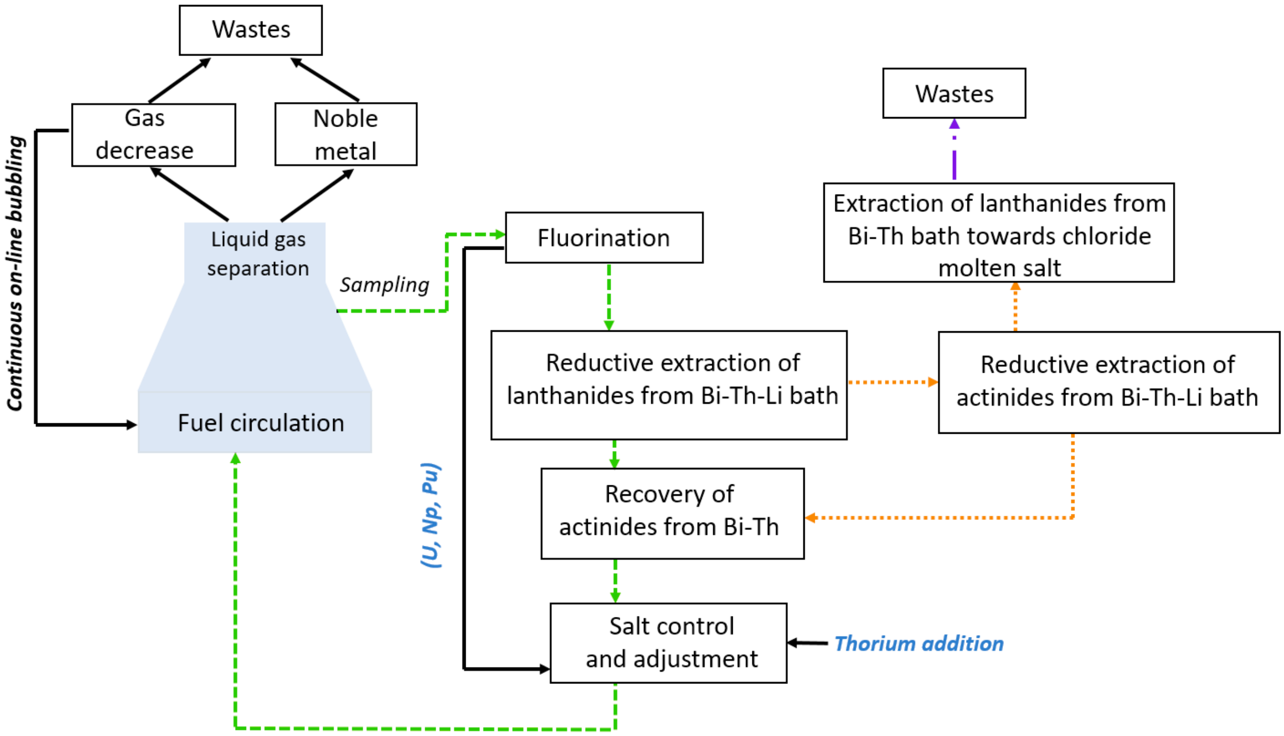

FIG. 18. Process flow and primary waste stream for Case 11 (MSR/MS/Py). Black arrows represent a gas stream, green arrows represent a fluoride stream; orange arrows show metallic bath handling and the purple arrow shows molten chloride use.

TABLE 9. SCREENING OF THE INNOVATIVE FUEL CYCLE OPTIONS

Reactor type	Fuel type	Reprocessing	Fuel fabrication	MA recycling	Remarks
SFR LFR (LMFR)	Oxide (U–Pu)	Aqueous	Pellet	○	Case 1 (FR/O/Aq)
		Pyro	Vibropac	×	Case 8 (FR/O/Py)
	Carbide (U–Pu)	Oxidation + aqueous	Pellet	○	Case 2 (FR/C/Aq)
	Nitride (U–Pu)	Aqueous	Conversion + pellet	○	Case 3 (FR/N/Aq)
		Pyro	Conversion + pellet	○	Case 9 (FR/N/Py)
	Metal (U–TRU)	Aqueous	×	○	Oxide to metal reduction may be less attractive
		Pyro	Injection casting	○	Case 10 (FR/M/Py)
VHTR	Oxide (U–Pu)	Crushing + aqueous	Sol-gel	○	Case 4 (VHTR/O/Aq)
		Crushing + pyro	×	×	No way to fabricate the coated particles
GCFR	Carbide (U–Pu)	Oxidation + aqueous	Sol-gel	○	Case 5 (GCFR/C/Aq)
	Nitride (U–Pu)	Oxidation + aqueous	Conversion + sol-gel	○	Case 6 (GCFR/N/Aq)
		Crushing + pyro	×	○	No way to fabricate the coated particles
MSFR	Molten fluoride salt Th–U–TRU	Pyro	Not necessary	Yes	Case 11 (MSR/MS/Py)
AHWR	Oxide (U–Th)	Aqueous	Pellet	Not necessary	Case 7 (AHWR/O/Aq)
		Pyro	×		DF may be too low for AHWR

Note: AHWR — advanced heavy water reactor; DF — dissolution of fuel; GCFR — gas cooled fast reactor; LFR — lead cooled fast reactor; LMFR — liquid metal fast reactor; MSFR — molten salt fast reactor; MSR — molten salt reactor; SFR — sodium cooled fast reactor; TRU — transuranic; VHTR — very high temperature reactor; ○ = no; × = yes.

TABLE 10. WASTE STREAMS FROM AQUEOUS REPROCESSING OF OXIDE FUEL FROM SFRs OR LFRs (CASE 1)

Component of waste	Stream description	Waste form	Technology status	Potential problems
Assembly hardware	Wrapper tube, nozzle, neutron shield and handling head	Cement, compaction? Ingot?	Industry stage Laboratory stage	H_2 gas generation by radiolysis needs to be addressed if immobilized in cement. More studies needed on disassembly method.
Hulls	Cladding and spacer wire	Cement, compaction, ingot Purification and reuse	Industry stage Pilot stage	H_2 gas generation by radiolysis needs to be addressed if immobilized in cement
Noble gas	Kr, Xe released (chopping and dissolution)	Released as gas waste or capture and decay store	Mature	Control the activity released
Iodine	Released as off-gas (chopping and dissolution)	Zeolite, ceramic matrix or release in liquid waste	Laboratory stage for conditioning	Iodine release in long term
^{14}C	Released in dissolution off-gas	Release with gaseous effluents, Scrubbed and released in liquid waste, carbonate waste form	Mature	^{14}C release in long term
NM[a]	Solids from the clarification	Glass form, ingot, or ingot with hulls	Industry stage, pilot stage	Specific glasses and process to be addressed
^{99}Tc	Liquid waste from extraction	Glass form with HLLW, or ingot with NM	Mature	
Cs/Sr	HLLW	Glass form	Mature	
Others	HLLW	Glass form	Mature	
Degraded solvent for major actinides	Degradation products in solvent washing solution, used solvent	Bitumen, vitrification, incineration	Mature	
Degraded solvent for minor actinides partitioning (optional)	Degradation products, used solvent (BTP, TODGA, etc.)	Ashes (combustion), cement? Ceramic?	Laboratory stage	Development of the solvent treatment (i.e. washing, regeneration, combustion/ conditioning are necessary)
Na-salt	Solvent washing, off-gas treatment	Glass form, cement	Mature	

TABLE 10. WASTE STREAMS FROM AQUEOUS REPROCESSING OF OXIDE FUEL FROM SFRs OR LFRs (CASE 1) (cont.)

Component of waste	Stream description	Waste form	Technology status	Potential problems
HNO_3	HNO_3 excess if operation is not balanced[b]	Release with LLW, cement after neutralization	Mature	
Combustible solid wastes	Paper scraps, gloves, etc.	Cement, compaction, ceramic or glass form? of ashes (combustion)	Mature	
Technological solid wastes	Equipment, piping, filters, etc.	Compaction, cement, ingot	Mature	
Decommissioning liquid waste	HLLW or ILLW from decontamination of equipment, piping, vessels	Cement, glass form after mineralization and concentration	Mature	
Decommissioning solid waste after decontamination	Equipment, vessels, piping, filters, etc.	Cement, compaction, ingot	Mature	

Note: HLLW — high level liquid waste; ILLW — intermediate level liquid waste; LFR — lead cooled fast reactor; LLW — low level waste; NM — noble metal; SFR — sodium cooled fast reactor.

[a] Noble metals such as Rh, Ru and Pd, often including Mo, Tc, Se and Te.

[b] There does not need to be excess HNO_3. Operations can be balanced to recycle HNO_3 and any excess goes with the HLW to vitrification where it’s converted to a mix of $N2_2$ and NO_x. The NO_x are generally recycled internally in the vitrification plant or destroyed in the off-gas treatment system. However, in some reprocessing systems, excess NO_3 may be needed as a neutron poison for criticality control purposes.

TABLE 11. WASTE STREAMS FROM AQUEOUS REPROCESSING OF CARBIDE FUEL FROM SFRs OR LFRs (CASE 2)

Component of waste	Stream description	Waste form	Technology status	Potential problems
Assembly hardware	Wrapper tube, nozzle, neutron shield and handling head	Cement, compaction? Ingot?	Industry stage Laboratory stage	H_2 gas generation by radiolysis needs to be addressed. More studies needed on disassembly method.
Hulls	Cladding and spacers (wires or grids)	Cement, compaction, ingot	Industry stage Pilot stage	H_2 gas generation by radiolysis needs to be addressed
Noble gas	Kr, Xe released (chopping and dissolution)	Released as gas waste	Mature	Control the activity released
Iodine	Released as off-gas from combustion and dissolution	Zeolite, ceramic matrix	Laboratory stage for conditioning	Iodine release in long term
^{14}C	Released as off-gas from combustion and dissolution	Scrubbed and released in liquid waste, carbonate waste form	Laboratory stage	Feasibility of the process should be confirmed by experiment ^{14}C release in long term
NM[a]	Solids from clarification	Glass form Or ingot with hulls	Industry stage, pilot stage	Specific glasses and process to be addressed
^{99}Tc	Liquid waste from extraction	Glass form with HLLW	Mature	
Cs/Sr	HLLW	Glass form	Mature	
Others	HLLW	Glass form	Mature	
Degraded solvent for major actinides	Degradation products in solvent washing solution, used solvent	Vitrification, incineration	Mature	
Degraded solvent for minor actinides partitioning (optional)	Degradation products, used solvent (BTP, TODGA, etc.)	Ashes (combustion), cement? ceramic?	Laboratory stage	Development of the solvent treatment (i.e. washing, regeneration, combustion/conditioning are necessary)
Na-salt	Solvent washing, off-gas treatment	Glass form, bitumen, cement	Mature	
HNO_3	HNO_3 excess	Release with LLW, cement after neutralization	Mature	

TABLE 11. WASTE STREAMS FROM AQUEOUS REPROCESSING OF CARBIDE FUEL FROM SFRs OR LFRs (CASE 2) (cont.)

Component of waste	Stream description	Waste form	Technology status	Potential problems
Combustible solid wastes	Paper scraps, gloves, etc.	Cement, compaction, ceramic or glass form? of ashes (combustion)	Mature	
Technological solid wastes	Equipment, piping, filters, etc.	Compaction, cement, ingot	Mature	
Decommissioning liquid waste	HLLW or ILLW from decontamination of equipment, piping, vessels	Cement, glass form after mineralization and concentration	Mature	
Decommissioning of solid waste after decontamination	Equipment, vessels, piping, filters, etc.	Cement, compaction, ingot	Mature	

Note: HLLW — high level liquid waste; ILLW — intermediate level liquid waste; LFR — lead cooled fast reactor; LLW — low level waste; SFR — sodium cooled fast reactor.

[a] Noble metals such as Rh, Ru and Pd.

TABLE 12. WASTE STREAMS FROM AQUEOUS REPROCESSING OF NITRIDE FUEL FROM SFRs OR LFRs (CASE 3)

Component of waste	Stream description	Waste form	Technology status	Potential problems
Assembly hardware	Wrapper tube, nozzle, neutron shield and handling head	Cement, compaction? Ingot?	Industry stage, laboratory stage	H_2 gas generation by radiolysis needs to be addressed. More studies needed on disassembly method.
Hulls	Cladding and spacers (wires or grids)	Cement, compaction, ingot	Industry stage, pilot stage	H_2 gas generation by radiolysis needs to be addressed
Noble gas	Kr, Xe released (chopping and dissolution)	Released as gas waste	Mature	Control the activity released
Iodine	Released as off-gas from combustion and dissolution	Zeolite, ceramic matrix	Laboratory stage for conditioning	Iodine release in long term
^{14}C	Released as off-gas from combustion and dissolution	Scrubbing and released in liquid waste, carbonate waste form	Laboratory stage	Need of enrichment of ^{15}N Development work needed to recover ^{15}N from the combustion off-gas ^{14}C release in long term
NM[a]	Solids from clarification	Glass form or ingot with hulls	Industry stage, pilot stage	Specific glasses and process to be addressed
^{99}Tc	Liquid waste from extraction	Glass form with HLLW	Mature	
Cs/Sr	HLLW	Glass form	Mature	
Others	HLLW	Glass form	Mature	
Degraded solvent for major actinides	Degradation products in solvent washing solution, used solvent	Vitrification, incineration	Mature	
Degraded solvent for minor actinides partitioning (optional)	Degradation products, used solvent (BTP, TODGA, etc.)	Ashes (combustion), cement? Ceramic?	Laboratory stage	Development of the solvent treatment (i.e. washing, regeneration, combustion/ conditioning are necessary)
Na-salt	Solvent washing, off-gas treatment	Glass form, bitumen, cement	Mature	

TABLE 12. WASTE STREAMS FROM AQUEOUS REPROCESSING OF NITRIDE FUEL FROM SFRs OR LFRs (CASE 3) (cont.)

Component of waste	Stream description	Waste form	Technology status	Potential problems
HNO_3	HNO_3 excess	Release with LLW, cement after neutralization	Mature	
Combustible solid wastes	Paper scraps, gloves, etc.	Cement, compaction, ceramic or glass form of ashes (combustion)	Mature	
Technological solid wastes	Equipment, piping, filters, etc.	Compaction, cement, ingot	Mature	
Decommissioning liquid waste	HLLW or ILLW from decontamination of equipment, piping, vessels	Cement, glass form after mineralization and concentration	Mature	
Decommissioning solid waste after decontamination	Equipment, vessels, piping, filters, etc.	Cement, compaction, ingot	Mature	

Note: HLLW — high level liquid waste; ILLW — intermediate level liquid waste; LFR — lead cooled fast reactor; LLW — low level waste; SFR — sodium cooled fast reactor.

[a] Noble metals such as Rh, Ru and Pd

TABLE 13. WASTE STREAMS FROM AQUEOUS REPROCESSING OF OXIDE FUEL FROM VHTRs (CASE 4)

Component of waste	Stream description	Waste form	Technology status	Potential problems
Assembly hardware	Prismatic block and graphite from the fuel compact/fuel spheres	Cement?	Conceptual stage	More studies are needed on disassembly method and conditioning
Coatings	Crushed coatings of the fuel particles Fragmented coatings by arc welding techniques	Cement? of graphite and SiC fragment	Conceptual stage	More studies are needed on crushing method of the coatings and conditioning
Noble gas	Kr, Xe released during crushing and dissolution of kernels	Released as gas waste	Mature	Control the activity released
^{3}H	Released as ^{3}H gas and HTO during crushing and dissolution of kernels	Released after dilution as gas and liquid waste, decay storing	Conceptual stage	Investigation of the distribution of ^{3}H in coated fuel particle
Iodine	Released as dissolution off-gas	Zeolite, ceramic matrix	Laboratory stage for conditioning	Iodine release in long term
^{14}C	Released as dissolution off-gas	Released in liquid waste or carbonate waste form after scrubbing	Mature	^{14}C release in long term
NM[a]	Solids from clarification	Glass form or ingot with hulls	Industry stage, pilot stage	Specific glasses and process to be addressed
^{99}Tc	Liquid waste from extraction	Glass form with HLLW	Mature	
Cs/Sr	HLLW	Glass form	Mature	Decay heat of package
Others	HLLW	Glass form	Mature	
Degraded solvent for major actinides	Degradation products in solvent washing solution, used solvent	Vitrification, incineration	Mature	
Degraded solvent for minor actinides partitioning (optional)	Degradation products, used solvent (BTP, TODGA, etc.)	Ashes (combustion), cement? ceramic?	Laboratory stage	Developmental of the solvent treatment (i.e. washing, regeneration, combustion/ conditioning are necessary)

TABLE 13. WASTE STREAMS FROM AQUEOUS REPROCESSING OF OXIDE FUEL FROM VHTRs (CASE 4) (cont.)

Component of waste	Stream description	Waste form	Technology status	Potential problems
Na-salt	Solvent washing, off-gas treatment	Glass form, bitumen, cement	Mature	
HNO_3	HNO_3 excess	Release with LLW, cement after neutralization	Mature	
Combustible solid wastes	Paper scraps, gloves, etc.	Cement, compaction, ceramic or glass form of ashes (combustion)	Mature	
Technological solid wastes	Equipment, piping, filters, etc.	Compaction, cement, ingot	Mature	
Decommissioning liquid waste	HLLW or ILLW from decontamination of equipment, piping, vessels	Cement, glass form after mineralization and concentration	Mature	
Decommissioning solid waste after decontamination	Equipment, vessels, piping, filters, etc.	Cement, compaction, ingot	Mature	

Note: HLLW — high level liquid waste; HTO — tritiated water; ILLW — intermediate level liquid waste; LLW — low level waste.

[a] Noble metals such as Rh, Ru and Pd.

TABLE 14. WASTE STREAMS FROM AQUEOUS REPROCESSING OF CARBIDE FUEL FROM GCFRs (CASE 5)

Component of waste	Stream description	Waste form	Technology status	Potential problems
Assembly hardware	SiC structure, and SiC grids	Cement? Ceramic matrix?	Conceptual stage	More studies are needed on disassembly method and conditioning
Noble gas	Kr, Xe released (chopping and dissolution)	Released as gas waste	Mature	Control the activity released
^{3}H	Release of ^{3}H gas and HTO from mechanical treatment and dissolution	Release as gas and liquid waste, decay storing	Conceptual stage	Investigation of the distribution of ^{3}H in the fuel and SiC structure is needed
Iodine	Released as off-gas from combustion and dissolution	Zeolite, ceramic matrix	Laboratory stage for conditioning	Iodine release in long term
^{14}C	Released as off-gas from combustion and dissolution	Scrubbed and released in liquid waste, carbonate waste form	Laboratory stage	Feasibility of the process should be confirmed by experiment. ^{14}C release in long term.
NM[a]	Solids from clarification	Glass form or ingot with hulls	Industry stage, pilot stage	Specific glasses and process to be addressed
^{99}Tc	Liquid waste from extraction	Glass form with HLLW	Mature	
Cs/Sr	HLLW	Glass form	Mature	
Others	HLLW	Glass form	Mature	
Degraded solvent for major actinides	Degradation products in solvent washing solution, used solvent	Vitrification, incineration	Mature	
Degraded solvent for minor actinides partitioning (optional)	Degradation products, used solvent (BTP, TODGA, etc.)	Ashes (combustion), cement? Ceramic?	Laboratory stage	Developmental of the solvent treatment (i.e. washing, regeneration, combustion/ conditioning are necessary)
Na-salt	Solvent washing, off-gas treatment	Glass form, bitumen, cement	Mature	
HNO_3	HNO_3 excess	Release with LLW, cement after neutralization	Mature	

TABLE 14. WASTE STREAMS FROM AQUEOUS REPROCESSING OF CARBIDE FUEL FROM GCFRs (CASE 5) (cont.)

Component of waste	Stream description	Waste form	Technology status	Potential problems
Combustible solid wastes	Paper scraps, gloves, etc.	Cement, compaction, ceramic or glass form of ashes (combustion)	Mature	
Technological solid wastes	Equipment, piping, filters, etc.	Compaction, cement, ingot	Mature	
Decommissioning liquid waste	HLLW or ILLW from decontamination of equipment, piping, vessels	Cement, glass form after mineralization and concentration	Mature	
Decommissioning solid waste after decontamination	Equipment, vessels, piping, filters, etc.	Cement, compaction, ingot	Mature	

Note: GCFR — gas cooled fast reactor; HLLW — high level liquid waste; HTO — tritiated water; ILLW — intermediate level liquid waste; LLW — low level waste.

[a] Noble metals such as Rh, Ru and Pd.

TABLE 15. WASTE STREAMS FROM AQUEOUS REPROCESSING OF NITRIDE FUEL FROM GCFRs (CASE 6)

Component of waste	Stream description	Waste form	Technology status	Potential problems
Assembly hardware	SiC structure and SiC grids	Cement? Ceramic matrix?	Conceptual stage	More studies are needed on disassembly method and conditioning
Noble gas	Kr, Xe released (chopping and dissolution)	Released as gas waste	Mature	Control the activity released
^{3}H	Release of ^{3}H gas and HTO from mechanical treatment and dissolution	Release as gas and liquid waste, decay storing	Conceptual stage	Investigation of the distribution of ^{3}H in the fuel and SiC structure is needed
Iodine	Released as off-gas from combustion and dissolution	Zeolite, ceramic matrix	Laboratory stage for the confining	Iodine release in long term.
^{14}C	Released as off-gas from combustion and dissolution	Scrubbed and released in liquid waste, carbonate waste form	Laboratory stage	Need of enrichment of ^{15}N Development work needed to recover ^{15}N from the combustion off-gas ^{14}C release in long term
NM[a]	Solids from clarification	Glass form or ingot with hulls	Industry stage, pilot stage	Specific glasses and process to be addressed
^{99}Tc	Liquid waste from extraction	Glass form with HLLW	Mature	
Cs/Sr	HLLW	Glass form	Mature	Decay heat of package
Others	HLLW	Glass form	Mature	
Degraded solvent for major actinides	Degradation products in solvent washing solution, used solvent	Vitrification, incineration	Mature	
Degraded solvent for minor actinides partitioning (optional)	Degradation products, used solvent (BTP, TODGA, etc.)	Ashes (combustion), cement? Ceramic?	Laboratory stage	Development of the solvent treatment (i.e. washing, regeneration, combustion/conditioning are necessary)
Na-salt	Solvent washing, off-gas treatment	Glass form, bitumen, cement	Mature	

TABLE 15. WASTE STREAMS FROM AQUEOUS REPROCESSING OF NITRIDE FUEL FROM GCFRs (CASE 6) (cont.)

Component of waste	Stream description	Waste form	Technology status	Potential problems
HNO_3	HNO_3 excess	Release with LLW, cement after neutralization	Mature	
Combustible solid wastes	Paper scraps, gloves, etc.	Cement, compaction, ceramic or glass form of ashes (combustion)	Mature	
Technological solid wastes	Equipment, piping, filters, etc.	Compaction, cement, ingot	Mature	
Decommissioning liquid waste	HLLW or ILLW from decontamination of equipment, piping, vessels	Cement, glass form after mineralization and concentration	Mature	
Decommissioning solid waste after decontamination	Equipment, vessels, piping, filters, etc.	Cement, compaction, ingot	Mature	

Note: GCFR — gas cooled fast reactor; HLLW — high level liquid waste; HTO — tritiated water; ILLW — intermediate level liquid waste; LLW — low level waste.

[a] Noble metals such as Rh, Ru and Pd.

TABLE 16. WASTE STREAMS FROM AQUEOUS REPROCESSING OF OXIDE FUEL FROM AHWRs (CASE 7)

Component of waste	Stream description	Waste form	Technology status	Potential problems
Assembly hardware	Nozzle and grids	Cement, compaction, ingot?	Industry stage, laboratory stage	H_2 gas generation by radiolysis needs to be addressed
Hulls	Cladding	Cement, compaction? Ingot?	Industry stage, pilot stage	H_2 gas generation by radiolysis needs to be addressed
Noble gas	Kr, Xe released (chopping and dissolution)	Released as gas waste	Mature	Control the activity released
Iodine	Released as off-gas (chopping and dissolution)	Zeolite, ceramic matrix	Laboratory stage for the conditioning	Iodine release in long term
^{14}C	Released in dissolution off-gas	Scrubbed and released in liquid waste, carbonate waste form	Mature	^{14}C release in long term
NM[a]	Solids from clarification	Glass form or ingot with hulls	Industry stage, pilot stage	Specific glasses and process to be addressed
^{99}Tc	Liquid waste from extraction	Glass form with HLLW	Mature	
Cs/Sr	HLLW	Glass form	Mature	
Others	HLLW	Glass form	Mature	
Degraded solvent for major actinides	Degradation products in solvent washing solution, used solvent	Vitrification, incineration	Mature	
Na-salt	Solvent washing, off-gas treatment	Glass form, bitumen, cement	Mature	
HNO_3	HNO_3 excess	Release with LLW, cement after neutralization	Mature	
Combustible solid wastes	Paper scraps, gloves, etc.	Cement, compaction, ceramic or glass form of ashes (combustion)	Mature	
Technological solid wastes	Equipment, piping, filters, etc.	Compaction, cement, ingot	Mature	

TABLE 16. WASTE STREAMS FROM AQUEOUS REPROCESSING OF OXIDE FUEL FROM AHWRs (CASE 7) (cont.)

Component of waste	Stream description	Waste form	Technology status	Potential problems
Decommissioning liquid waste	HLLW or ILLW from decontamination of equipment, piping, vessels	Cement, glass form after mineralization and concentration	Mature	
Decommissioning solid waste after decontamination	Equipment, vessels, piping, filters, etc.	Cement, compaction, ingot	Mature	

Note: HLLW — high level liquid waste; ILLW — intermediate level liquid waste; LLW — low level waste.

[a] Noble metals such as Rh, Ru and Pd.

TABLE 17. WASTE STREAMS FROM PYRO REPROCESSING OF OXIDE FUEL FROM SFRs OR LFRs (CASE 8)

Component of waste	Stream description	Waste form	Technology status	Potential problems
Assembly hardware	Wrapper tube, nozzle, neutron shield, bottom head and handling head	Cement, compaction? Ingot?	Industry stage, laboratory stage	H_2 gas generation by radiolysis needs to be addressed. More studies needed on disassembly method.
Hulls	Cladding and spacers (wires or grids)	Cement, compaction, ingot	Industry stage, pilot stage	H_2 gas generation by radiolysis needs to be addressed.
Noble gas	Kr, Xe released (chopping and dissolution in molten salt)	Released as gas waste Capture and store	Pilot stage Conceptual stage	More studies are needed to separate noble gas from the chlorine off-gas, such as the cryogenic process.
Iodine	Mixed in molten salt or chlorine gas	Phosphate glass, sodalite, ceramic matrix	Conceptual stage	More studies are needed to separate iodine from the chlorine off-gas, such as the cryogenic process, to develop conditioning process. Iodine release in long term.
^{14}C	Mixed in molten salt as solid or chlorine gas as CO_2 or CCl_4 gas	Released as gas waste, carbonate waste form	Conceptual stage	More studies are needed to separate ^{14}C from the chlorine off-gas such as the cryogenic process, to develop conditioning process. ^{14}C release in long term.
NM	Cathode deposit during electro-winning step	Ingot	Laboratory stage	Development work is needed regarding alloying of NM in high temperature.
^{99}Tc	Cathode deposit during electro-winning step	Ingot	Laboratory stage	Development work is needed regarding alloying of NM in high temperature.
Cs/Sr	Molten salt	Glass form (after oxide conversion in high temperature with H_3BO_3) Sodalite	Conceptual stage, pilot stage	More studies are needed regarding the behaviour of Cs/Sr chloride during the conversion.
Others	Solid phosphate precipitate from molten salt or electrodeposition	Phosphate glass form Sodalite waste form	Laboratory stage Laboratory stage	Developmental work on vitrification process.

TABLE 17. WASTE STREAMS FROM PYRO REPROCESSING OF OXIDE FUEL FROM SFRs OR LFRs (CASE 8) (cont.)

Component of waste	Stream description	Waste form	Technology status	Potential problems
Solvent	Molten salt	Glass form (after oxide conversion in high temperature with H_3BO_3) Sodalite	Conceptual stage Pilot stage	More studies are needed regarding the behaviour of chloride salt during the conversion.
Combustible solid wastes	Paper scraps, gloves, etc.	Cement, compaction, ceramic or glass form of ashes (combustion)	Mature	
Technological solid wastes	Equipment, piping, filters, etc.	Compaction, cement, ingot	Mature	
Decommissioning liquid waste	HLLW or ILLW from decontamination of equipment, piping, vessels	Cement, glass form after mineralization and concentration	Mature	
Decommissioning solid waste after decontamination	Equipment, vessels, piping, filters, etc.	Cement, compaction, ingot	Mature	

Note: HLLW — high level liquid waste; ILLW — intermediate level liquid waste; LFR — lead cooled fast reactor; NM — noble metal; SFR — sodium cooled fast reactor.

[a] Noble metals such as Rh, Ru, Pd, Fe, Zr, Tc, Mo, etc. (everything more noble than U in alkali chloride salts).

TABLE 18. WASTE STREAMS FROM PYRO REPROCESSING OF NITRIDE FUEL FROM SFRs OR LFRs (CASE 9)

Component of waste	Stream description	Waste form	Technology status	Potential problems
Assembly hardware	Wrapper tube, nozzle, neutron shield and handling head	Cement, compaction, ingot?	Industry stage, laboratory stage	H_2 gas generation by radiolysis needs to be addressed. More studies needed on disassembly method.
Hulls	Cladding and spacers (wires or grids)	Ingot	Pilot stage	H_2 gas generation by radiolysis needs to be addressed.
Noble gas	Kr, Xe released (chopping and dissolution in molten salt)	Released as gas waste	Industry stage	Buildup of noble gas concentration in Ar gas needs to be addressed.
Iodine	Mixed in molten salt or Ar off-gas	Zeolite, ceramic matrix	Conceptual stage	More studies are needed regarding behaviour of Iodine during the Sodalite production. Amount of Ar gas purification column material needs to be addressed. Iodine release in long term.
^{14}C	Mixed in molten salt or Ar off-gas	Zeolite, ceramic matrix	Conceptual stage	More studies are needed regarding the behaviour of carbon in the electro-refining process. ^{14}C release in long term.
NM[a]	Anodic residues and Cd pool	Ingot	Pilot stage	The effect of adhered salt and Cd during the induction heating needs to be addressed.
^{99}Tc	Anodic residues and Cd pool	Ingot	Pilot stage	The effect of adhered salt and Cd during the induction heating needs to be addressed.
Cs/Sr	Molten salt waste	Sodalite	Pilot stage	Reduction of the waste volume needs to be addressed. Long term behaviour.
Others	Molten salt waste	Sodalite	Pilot stage	Reduction of the waste volume needs to be addressed. Long term behaviour.
Solvent	Molten salt	Sodalite	Pilot stage	Reduction of the waste volume needs to be addressed. Long term behaviour.

TABLE 18. WASTE STREAMS FROM PYRO REPROCESSING OF NITRIDE FUEL FROM SFRs OR LFRs (CASE 9) (cont.)

Component of waste	Stream description	Waste form	Technology status	Potential problems
^{15}N	Released ^{15}N (dissolution)	Lost ^{15}N released with Ar as gas waste form	Laboratory stage	Feasibility of ^{15}N collection should be confirmed by experiment.
Combustible solid wastes	Paper scraps, gloves, etc.	Cement, compaction, ceramic or glass form of ashes (combustion)	Mature	
Technological solid wastes	Equipment, piping, filters, etc.	Compaction, cement, ingot	Mature	
Decommissioning liquid waste	HLLW or ILLW from decontamination of equipment, piping, vessels	Cement, glass form after mineralization and concentration	Mature	
Decommissioning solid waste after decontamination	Equipment, vessels, piping, filters, etc.	Cement, compaction, ingot	Mature	

Note: HLLW — high level liquid waste; ILLW — intermediate level liquid waste; LFR — lead cooled fast reactor; NM — noble metal; SFR — sodium cooled fast reactor.

[a] Noble metals such as Rh, Ru and Pd.

TABLE 19. WASTE STREAMS FROM PYRO REPROCESSING OF METALLIC FUEL FROM SFRs OR LFRs (CASE 10)

Component of waste	Stream description	Waste form	Technology status	Potential problems
Assembly hardware	Wrapper tube, nozzle, neutron shield and handling head	Cement, compaction? Ingot?	Industry stage, laboratory stage	H_2 gas generation by radiolysis. More studies needed on disassembly method.
Hulls	Cladding and spacers (wires or grids)	Ingot with the NM	Pilot stage	
Noble gas	Kr, Xe released (chopping and dissolution in molten salt)	Released as gas waste	Pilot stage	Buildup of noble gas concentration in Ar gas needs to be addressed.
^{3}H	?	Decay storing as liquid waste?	?	Investigation on the distribution of ^{3}H in metallic fuel is needed.
Iodine	Mixed in molten salt or Ar off-gas	Zeolite, ceramic matrix	Conceptual stage	More studies are needed regarding behaviour of iodine during the sodalite production. The amount of column material for the Ar gas purification needs to be addressed. Iodine release in long term.
^{14}C	Mixed in molten salt or Ar off-gas	Release in off-gas, carbonate waste form	Conceptual stage	More studies are needed regarding the behaviour of carbon in the electro-refining process. ^{14}C release in long term.
NM[a]	Anodic residues and Cd pool	Ingot with the hulls	Pilot stage	The effect of adhered salt and Cd during the induction heating needs to be addressed.
^{99}Tc	Anodic residues and Cd pool	Ingot	Pilot stage	The effect of adhered salt and Cd during the induction heating needs to be addressed.
Cs/Sr	Molten salt waste	Sodalite	Pilot stage	Reduction of the waste volume needs to be addressed. Long term behaviour.

TABLE 19. WASTE STREAMS FROM PYRO REPROCESSING OF METALLIC FUEL FROM SFRs OR LFRs (CASE 10) (cont.)

Component of waste	Stream description	Waste form	Technology status	Potential problems
Others	Molten salt waste	Sodalite	Pilot stage	Reduction of the waste volume needs to be addressed. Long term behaviour.
Solvent	Molten salt	Sodalite	Pilot stage	Reduction of the waste volume needs to be addressed. Long term behaviour.
Bonding sodium	Molten salt or Na recovery prior to anodic dissolution	Sodalite Sodalite (added as oxide)	Pilot stage, laboratory stage	
Silica mould	Injection casting process	Cement Sodalite	Pilot stage, laboratory stage	
Combustible solid wastes	Paper scraps, gloves, etc.	Cement, compaction, ceramic or glass form of ashes (combustion)	Mature	
Technological solid wastes	Equipment, piping, filters, etc.	Compaction, cement, ingot	Mature	
Decommissioning liquid waste	HLLW or ILLW from decontamination of equipment, piping, vessels	Cement, glass form after mineralization and concentration	Mature	
Decommissioning solid waste after decontamination	Equipment, vessels, piping, filters, etc.	Cement, compaction, ingot	Mature	

Note: HLLW — high level liquid waste; ILLW — intermediate level liquid waste; LFR — lead cooled fast reactor; NM — noble metal; SFR — sodium cooled fast reactor.

[a] Noble metals such as Rh, Ru and Pd often include Mo, Tc, Se and Te.

TABLE 20. WASTE STREAMS FROM PYRO REPROCESSING OF MSR FUEL (CASE 11)

Component of waste	Stream description	Waste form	Technology status	Potential problems
Noble gas	Kr, Xe off-gas in dilution with inert gas sent to storage for decay into Rb, Cs, Sr, Ba. Recycling of the gas mixture after a few hours of storage.	Medium term gas storage (100 years) Metallic waste in Pb	Industry stage	Buildup of noble gas concentration in Ar gas needs to be addressed.
^{3}H	Distribution not known between off-gas and molten salt. Extracted from the salt at the fluorination step for U extraction (HF).	Tritiated water	Conceptual stage	Investigation of the distribution of ^{3}H between off-gas and molten salt is needed.
Iodine	Mixed in molten salt because of redox potential control in the fuel and extracted by HF or F_2	Zeolite, ceramic matrix, sodalite (with molten salt)	Laboratory stage	Development of the conditioning process is needed. Iodine release in long term.
^{14}C	Not present in MSFR as moderator Present as solid ceramics (SiC, B_4C)	Carbonate waste form	Conceptual stage	More studies are needed regarding the behaviour of carbon and its waste form.
NM[a]	Solid metallic particles in molten salt or off-gas processing	Ingot	Conceptual stage	More studies are needed regarding the behaviour of NM and its waste form.
^{99}Tc	Alloyed with NM	Glass form, ingot	Conceptual stage	
Cs/Sr	Molten salt or metallic form in noble gas decay storage	Glass or metallic form	Conceptual stage	Development of the conditioning process is needed. Long term behaviour.
Others	Extraction from the salt and converted into oxide (lanthanides, Zr) or associated with NM	Fluoride glass form	Laboratory stage	Development of the conditioning process is needed. Long term behaviour.
Solvent	Recycled	Fluoride glass form	Laboratory stage	Development of the conditioning process is needed. Long term behaviour.
Combustible solid wastes	Paper scraps, gloves, etc.	Cement, compaction, ceramic or glass form of ashes (combustion)	Mature	

TABLE 20. WASTE STREAMS FROM PYRO REPROCESSING OF MSR FUEL (CASE 11) (cont.)

Component of waste	Stream description	Waste form	Technology status	Potential problems
Technological solid wastes	Equipment, piping, filters, etc.	Compaction, cement, ingot	Mature	
Decommissioning liquid waste	HLLW or ILLW from decontamination of equipment, piping, vessels	Bitumen, cement, glass form after mineralization and concentration	Mature	
Decommissioning of solid waste after decontamination	Equipment, vessels, piping, filters, etc.	Cement, compaction, ingot	Mature	

Note: HLLW — high level liquid waste; ILLW — intermediate level liquid waste; MSFR — molten salt fast reactor; MSR — molten salt reactor; NM — noble metal.

[a] Noble metals such as Rh, Ru and Pd often include Mo, Tc, Se and Te.

4.9. MANAGEMENT OF WASTE

The waste from fuel cycle facilities may contain [24, 27, 32]:

- High concentrations of metals such as Zr, Cr, Ni, Fe, Al and their nitrates (e.g. from fuel cladding, hulls and other hardware);
- Alkali metal fluorides or chlorides;
- Uranium contaminated compounds or metals.

While the waste streams may be similar for different fuel types for both aqueous and pyro processing methods, the quantities produced and their actual characteristics may differ for each of the fuel types. In addition, although the process is similar in principle for the different fuel types, the actual plant configuration and operation will be specific to an individual fuel type. This may affect the chemical properties of the waste stream and the resultant form of the waste products. Plant configuration will also affect maintenance waste, as well as waste from future decommissioning.

4.9.1. Solid waste

As described in Section 4.3, a wide variety of technologies are available for treatment and disposal of L&ILW. These technologies can be applied to L&ILW generated at fuel cycle facilities.

The management of cladding hulls is an important issue for long term sustainability. Low burnup reactors require more cladding hulls than reactors with higher burnup fuels, due to the larger quantity of fuel required to achieve the same net energy output. Although this waste may not be heat generating, it will have a very high radiation field due to induced activity and contain adhered fuels, making it unsuitable for disposal in near surface disposal facilities.

At present, compaction is the most widely adopted practice for managing metallic hardware; however, volume reduction by this technique is modest. Eutectic melting could reduce the volume considerably and needs to be studied further. Also, in the case of zirconium based cladding, purification of zirconium for recycling or LLW disposal should be studied.

4.9.2. Liquid waste

In reprocessing plants, extraction of fissile/fertile materials (i.e. uranium, plutonium and thorium) is well established.

Recovery of minor actinides and long lived fission products from HLLW is essential for optimizing the waste management steps and ensuring that immobilized waste need not be monitored for very long periods. Also, effective recovery of the actinides and fission products would permit a significant reduction in repository space (assuming that caesium/strontium are managed without repository disposal), which could otherwise be a limiting factor for the growth of reactor systems.

In the late 1980s, studies at the Argonne National Laboratory (ANL) resulted in the development of a unique extractant — n-octyl, phenyl N, N–di-isobutyl carbamoylmethyl phosphine oxide (usually referred to as CMPO) for the recovery of actinides from HLLW solutions. This extractant can be used to recover actinides from high level aqueous waste generated from thermal and fast reactor fuel reprocessing. Flowsheets based on CMPO have been developed and demonstrated on a pilot plant scale in European countries and elsewhere. However, to date, none of the operating reprocessing plants in the world have integrated minor actinide partitioning with the main flowsheet for reprocessing. Newer extractant systems such as di-glycolamides have since emerged, promising better extraction characteristics; however, experience with these extractants is limited to lab scale experiments, and thus far has been almost entirely restricted to the processing of inactive streams. Indeed, radiolytic decomposition of solvents and extractant molecules is a problem that needs to be overcome or controlled.

Separation of actinides from lanthanides in the mixture obtained by the separation processes described above is another area where work is still in the laboratory domain. Several extractants (such as HDEHP, BTP, Cyanex 301) have been considered as candidates, but there is no consensus on which would be the most suitable extractant. Also, there has been no techno-economical evaluation on the separation of minor actinides and their subsequent utilization. Virtually no work has been done to understand the washing processes and their impacts on waste management.

It is well known that the HLLW solution resulting from reprocessing of fast reactor fuels contains a significant amount of isotopes such as ^{137}Cs and ^{90}Sr that could be used for non-nuclear applications. For example, the Russian Federation routinely recovers large quantities of ^{137}Cs. However, there have been no reports of recovery of these isotopes from irradiated fast reactor fuels. It is conceivable that the elements of waste management that involve recovery of minor actinides and fission products are built into flowsheets for fuel reprocessing.

The solvent extraction and ion exchange processes used in reprocessing generate significant quantities of aqueous wastes. Reduction in the number of cycles of purification is one approach to addressing this issue. This would be aided by adopting radiation tolerant extractants that promise higher decontamination factors from fission products. Another issue concerns reduction of alpha bearing solid waste by suitable decontamination techniques without generating secondary wastes. The existing processes also use organic reagents (e.g. N-dodcane as the diluent in PUREX process) which have to be disposed of after repeated use. Further R&D is needed to avoid the use of reagents that add to the salt content of the waste solutions. Photochemical and electrochemical processes could address this issue.

In addition, there is a need to develop waste immobilization matrices that can tolerate a higher salt content and remain stable for extended periods in a geological environment, especially for fast reactor fuel cycles. While ceramic matrices for waste immobilization have been under development in many countries, none has been applied on an industrial scale. Innovations are needed to identify ceramic matrices with a lower process temperature and tolerance for higher loading in terms of salt content and radioactivity content. Ceramic immobilization could represent an emerging alternative or a complement to current vitrification.

A titanate based synthetic rock (synroc) is the most widely studied crystalline waste form. The candidate mineral phases in any synroc type assemblage usually have relatively complex crystal structures with polyhedral coordination of different sizes and shapes; this provides different substitution schemes with a charge balance for elements in the HLLW, with or without minor changes in the crystal structure. The principal minerals of the synroc assemblage can each accommodate a range of radionuclides. Typically, hollandite ($BaAl_2Ti_6O_{16}$) can incorporate elements such as caesium, rubidium and barium; zirconolite ($CaZrTi_2O_7$) can host uranium, zirconium, neptunium, plutonium and rare earths; perovskite ($CaTiO_3$) can take up strontium, neptunium and plutonium. Rutile helps in the microencapsulation of the minor alloy phases and also increases the mechanical strength of the waste form. In some formulations, pyrochlore ($A_2B_2O_7$, in which A and B are cations), which has a crystal structure closely related to the zirconolite structure, may become the actinide host phase. The mineral phases formed, and their relative amounts, are determined by the additives. Inexpensive and robust processes to form the ceramic waste forms need to be developed and demonstrated.

With the advances in waste partitioning, ceramic matrices have become an attractive proposition. Durable single phase matrices can be designed to immobilize particular waste streams containing one element or a group of chemically similar radionuclides.

5. DECOMMISSIONING WASTE

Planning for decommissioning of innovative reactors and fuel cycle facilities is an important aspect that cannot be overlooked in the design process. Often, engineering choices made during the design stage have a profound impact on later decommissioning of those facilities. These need to be carefully considered up front as part of an integrated waste management system in order to avoid complications later. For example:

- Choice of materials in the reactor core will influence the radionuclides created through neutron activation, as well as the amount and characteristics of surface contamination that will be deposited on the internal surfaces of equipment, ex-core;
- Choice of building materials for potentially contaminated areas will influence the ease and method of decontamination, as well as the amount of secondary wastes;

— Ease of access to equipment will determine dismantling methods, which will influence worker dose and the waste produced by the conduct during the decommissioning operations.

Equipment lists, construction plans and as-built construction documentation all serve as useful information for planning and executing a decommissioning project. Such documentation needs to be preserved as part of the overall decommissioning plan [33].

A decommissioning strategy can be developed based on existing policy in the Member State (e.g. prompt vs. delayed decommissioning). Next, a conceptual decommissioning plan can be developed. For conservatism, it should be based on the use of current technology for dismantling and waste management. The plant equipment lists, with their locations in the plant and descriptions of how they are used, are a good starting point for developing the decommissioning waste stream list, which must include items such as estimated quantities, required treatment and conditioning methods. Some standard decommissioning planning was documented in numerous other reports (see Refs [33–37]). The same principles of decommissioning planning apply to both innovative reactors and their associated fuel cycle facilities.

The unique aspects of innovative nuclear reactors and fuel cycle facilities relate to their use of exotic materials in construction, non-aqueous coolants (e.g. liquid metals) and the complex chemical separation processes used in fuel preparation and reprocessing, and to the high intensity and hard spectra of neutron flux in the core. All of these factors will generate waste materials during decommissioning that are not produced by current facilities.

5.1. WASTE FROM DECOMMISSIONING OF INNOVATIVE NUCLEAR REACTORS

Waste from decommissioning of innovative reactors is initiated when an operating nuclear reactor is permanently shut down. The waste results from the removal of fuel and coolant, the dismantling of the reactor structure, the containment and associated buildings, including the major components (such as the gas manifold, neutron shield, graphite moderator, internal support structure and the pressure vessel itself), and remote handling equipment (such as manipulators, hoists, transfer and viewing equipment), as applicable to the specific design of the facility. Most of the decommissioning wastes from innovative reactors are similar to the waste from conventional reactors. However, wastes from the coolant and reactor core are different.

A summary of waste from the decommissioning of innovative reactors is shown in Table 21. Further details can be found in the reactor waste stream lists in Section 4. While not all of these new waste streams will be problematic, each waste stream needs to be quantified and examined on a case by case basis to determine a suitable management method and final disposition. The volumes and characteristics of the waste will depend on the specific reactor design. The characteristics will also depend on the operating history of the reactor (e.g. irradiation flux and time, fuel leakage history, maintenance history).

5.2. WASTE FROM DECOMMISSIONING OF INNOVATIVE FUEL CYCLE FACILITIES

Table 22 summarizes the principal waste streams from the decommissioning of fuel cycle facilities. For fuel cycle facilities, the major sources of waste during decommissioning result from the disassembly of process equipment, hot cells, remote handling equipment, storage tanks, waste processing areas (e.g. vitrification systems) and associated buildings and structures. The fuel cycle facilities will be dominated by surface contamination (instead of direct activation by neutron flux as in the reactor core), but will also contain residues of harsh and corrosive chemicals used in reprocessing operations, and elevated levels of alpha-emitting radionuclides.

Generally, unnecessary clean equipment is removed first. (Some equipment, such as air, electrical, ventilation, water and waste systems may still be required in order to support the decommissioning operations.) Dismantling of the radioactive areas involves a series of steps, one for each of the major components (e.g. gas manifold, neutron shield, graphite moderator, internal support structure and the pressure vessel itself). Generally, steps proceed from ‘cleaner’ areas to more contaminated areas to avoid the risk of contaminating equipment in the clean areas. Remote handling equipment and other support systems that may not already be present, including manipulators, hoists, transfer and viewing equipment, need to be installed for decommissioning. In addition, a waste monitoring system for testing, encapsulation and shielded boxes for on-site storage may be necessary. Resulting wastes need to be

TABLE 21. SUMMARY OF WASTE FROM THE DECOMMISSIONING OF INNOVATIVE REACTORS

Reactor type	Routine waste	Unique waste	Comments
SFR	Dismantled components and structures	Sodium wetted combustible waste Sodium wetted metallic waste Bulk sodium from cooling system	Several sodium cooled reactors are under decommissioning and three of them have been completed (including sodium cooled moderate reactors). Special processes have been developed to manage sodium contaminated materials which are treated as waste.
GCFR	Dismantled components and structures	Tritium embrittled steel	No significant problems expected
LFR	Dismantled components and structures	Lead and bismuth contaminated materials Bulk lead and bismuth from cooling system	Simple processes have already been developed to manage lead (and bismuth) wetted materials, treated as waste.[a] Standard provisions of conventional industry are required to manage lead vapours and aerosols.
SCWR	Dismantled components and structures	No unique waste expected	No significant problems expected
AHWR	Dismantled components and structures	No unique waste expected	No significant problems expected
VHTR	Dismantled components and structures	Large volume of waste graphite as LLW	Special process to fix graphite dust during decommissioning, increasing the volume of waste
MSR	Dismantled components and structures	Fluoride salt containing actinides and soluble fission products Metallic particles	The pyrochemical treatments of the salts or metallic fission products are still mostly at a conceptual stage, even if some steps were operated at laboratory scale in the past.

Note: AHWR — advanced heavy water reactor; GCFR — gas cooled fast reactor; LFR — lead cooled fast reactor; LLW — low level waste; MSR — molten salt reactor; SCWR — supercritical water cooled reactor; SFR — sodium cooled fast reactor; VHTR — very high temperature reactor.

[a] Cleaning Methods for LFR/SFR Components, ENEA Report NNFISS, LP3 – 021 2/9/2011 (public, in Italian).

TABLE 22. SUMMARY OF WASTE FROM DECOMMISSIONING OF INNOVATIVE FUEL CYCLE FACILITIES

Facility type	Routine waste	Unique waste	Comments
Enrichment	Dismantled components and structures	No change to current facilities	
Conversion	Dismantled components and structures	Chemical wetted process equipment, sludges in tanks	
Fuel fabrication	Dismantled components and structures	Exotic materials, equipment with high levels of residual actinides	
Aqueous reprocessing	Dismantled components and structures	Chemical wetted process equipment, sludges in tanks, exotic materials, equipment with high levels of fission products and residual actinides	
Pyro reprocessing	Dismantled components and structures	Same as MSFR fuel: Fuel salt contaminated or activated structure material (equipment with high levels of fission products and residual actinides). Liquid metal (Bi, Pb). Chloride salt from lanthanide partitioning.	
Liquid HLW treatment	Dismantled components and structures	Chemical wetted process equipment, sludges in tanks, exotic materials, equipment with high levels of fission products and residual actinides	

Note: HLW — high level waste; MSFR — molten salt fast reactor.

TABLE 23. COMMON WASTE STREAMS FROM DECOMMISSIONING OF FUEL CYCLE REPROCESSING FACILITIES

Typical source of waste	Waste stream	Waste form	Technology status	Potential problems
Cell lining	Decontaminated cell linings will be dismantled and cemented in a can. Categorized to transuranic (TRU), high active and low active waste.	Cement	Mature	
Containment box	Decontaminated containment boxes will be dismantled and cemented in a can. Categorized to TRU, high active and low active waste.	Cement	Mature	
Contaminated concrete	Contaminated concrete will be ground from the cell surface to be cemented.	Cement	Mature	
Out of cell equipment	Decontaminated equipment will be dismantled and cemented in a can. Categorized to low active waste and clearance level waste.	Cement/clearance level	Mature	
Clean concrete	Clean concrete will be separated from inner reinforced steel bars and disposed of as clearance level waste.	Clearance level waste	Mature	
Clean metal	Separated reinforce steel bars will be recycled.	Recycled	Mature	

classified for disposal at an appropriate facility (e.g. LLW, ILW, HLW). After all radioactive equipment is removed, the building areas can be decontaminated and dismantled or reused, if necessary.

Wastes from the decommissioning of reprocessing plants are more or less similar to those from current facilities. However, some degree of processing and/or conditioning will generally be required in order to meet the waste acceptance criteria for the receiving facility. Wastes from contaminated equipment, piping and vessels installed in cells could vary in each case and are described within Tables 9 to 16. New products of organic compounds (gel and mousse) supporting corrosive reagents like cerium (IV) are very efficient to decrease the dose rates of stainless steel equipment contaminated with actinides; these are in the industrialization stage. The remaining wastes arising, such as building concrete, are common for all facilities and are summarized in Table 23.

6. SUMMARY OF WASTE MANAGEMENT ISSUES

Table 24 summarizes the fuels and materials that may be used in innovative reactor systems. Some innovative reactor systems discussed in this report operate with temperatures that are higher than current nuclear industry experience. Also, all systems require longer service lives and high burnup for fuels. This, combined with the fact that they use fast or epithermal neutron spectra and often use very aggressive coolants (e.g. molten metals or molten salts), means they will require the use of exotic materials such as refractory alloys and ceramics. This may introduce activation products not currently seen in the nuclear industry, or introduce them in larger amounts than in previous reactor systems. In some cases, the use of a new type of fuel (e.g. nitride, carbides) will result in new waste streams. The use of these different materials and their activation products will also have an impact on the amount and type of operational and plant decommissioning waste produced by the different reactor types. While it is too early to state with certainty which materials will be used in the various reactor systems, the following sections and summary tables list the materials currently considered and some factors that should be considered in the waste management scheme.

Table 25 summarizes some of the reactor waste streams that may require special attention during the design, development and deployment of innovative reactor systems. In these cases, it is expected that technology can be developed to manage these wastes safely. The concern is that the waste is substantially different from the wastes currently seen from water or gas cooled NPPs and may require special considerations (e.g. sodium contamination, or the impact of fuel laden molten salts). Table 26 summarizes some of the waste streams from innovative fuel cycles that may require special development. The concern is that additional R&D may be required to adapt the fuel reprocessing techniques to the new fuel types and to develop suitable immobilization matrices for the resulting waste streams. Until this research is carried out, the form, nature and volume of the waste will not be known with any degree of certainty. The text that follows indicates findings related to fuel cycle waste, operational waste and decommissioning waste of innovative systems that are being considered.

Fuel cycle waste — uranium/plutonium

Figure 19 indicates differences in timescales required for the radiotoxicity to reach reference levels (natural uranium) in three development scenarios for a uranium fuel cycle (open cycle, full fissile recycling and full actinide recycling). In these recycling schemes, the radiotoxicity of HLW can be reduced by a factor of 100 to 200 and the timescale can be reduced from over 100 000 years to less than 1000 years by the introduction of a closed fuel cycle and fast reactors in a first step to recycle uranium/plutonium, and in a second step to burn minor actinides. Both steps would require the introduction of innovative reactor and fuel cycle concepts. For the first step, the most challenging task is probably developing new matrices for immobilization of HLW. In the later step, the most challenging aspect is developing dedicated actinide burners (FR, MSR and/or ADS) and applying new reprocessing schemes [1]. It is to be noted that radiotoxicity doesn't correlate at all to the projected disposal system performance criteria (e.g. dose to humans).

Further research is needed to develop waste treatment and conditioning technologies suitable for all potential waste streams. This work should continue in parallel with the development of the reactor systems themselves, such that the waste treatment and conditioning systems are ready for deployment at the same time as the reactor systems.

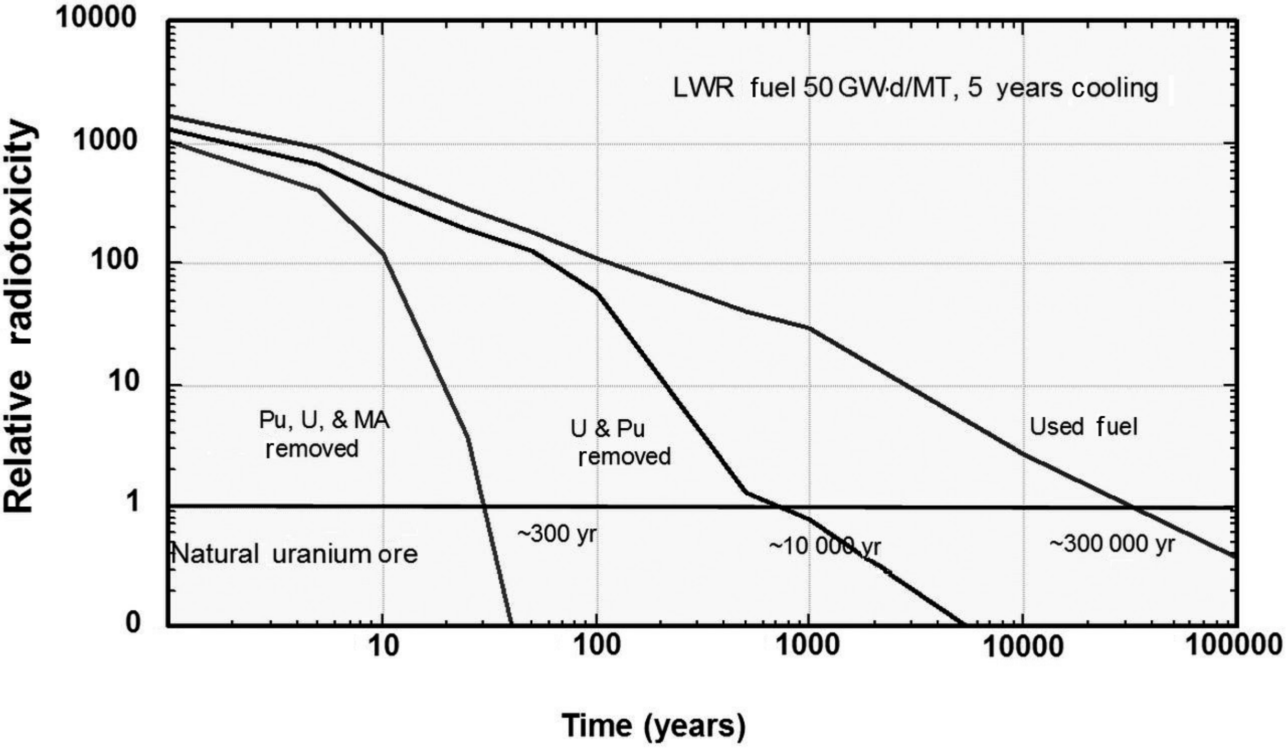

FIG. 19. *Relative radiotoxicity over time. LWR — light water reactor.*

With respect to the development of new matrices for HLW arising from the recycling of uranium/plutonium in fast reactors, the volume of high level aqueous waste produced in reprocessing is mainly caused by the fission product content, which again is limited by the achievable loading in the matrix for waste immobilization. With respect to a fast reactor fuel cycle, there is an incentive to develop waste matrices that can tolerate a higher salt content and can also remain stable for extended periods in a geological environment. Glass and ceramic matrices for waste immobilization have been under development in many countries for this purpose. Identifying matrices with a tolerance for higher loading in terms of salt content as well as radioactivity content is an important innovation. Studies on radionuclide migration in the environment, development of stable waste forms, and models that could reliably predict the long term behaviour of waste forms will continue to be important topics of research.

Research on utilization and transmutation of the minor actinides and long lived fission products has led to very different concepts from the standpoint of the reactors themselves, as well as the fuel and targets, and the corresponding scenarios for nuclear reactor deployment. New developments in spent fuel reprocessing should be more resistant to irradiation effects, and are therefore suitable for use with concentrated media. This is a further incentive to develop pyrochemical processing concepts in order to confirm and better assess the expected advantages. However, aqueous processes, due to their inherent advantages (high recovery efficiencies and well established technical maturity), remain at the forefront of reprocessing technology. Many improvements to the technology have been made over the years.

While fabrication of uranium and plutonium fuel is well established, there is limited experience available for fuel containing high levels of minor actinides and ^{238}Pu. The basic problem is associated with the high alpha activity and heat. This means fuel fabrication has to be carried out in alpha tight hot cells with adequate shielding. Better understanding is needed on the lanthanide concentration and burnup limits for fuels containing minor actinides before significant fuel cladding interactions occur. Advancements in fuel fabrication techniques are also needed.

Partitioning of minor actinides followed by waste conditioning into a very stable matrix appears to be the most appropriate method to improve current waste disposal options and reduce the long term risk. The application of partitioning and transmutation would, if fully implemented, result in a significant decrease in the TRU inventory to be disposed of in geological repositories.

Fuel cycle thorium (solid and liquid fuel)

Embarking on a reactor programme with thorium fuel may necessitate the development of new materials, alloys and more efficient solvents for extraction, and better equipment design for short contact time to minimize solvent degradation. The management of HLW from such reactors involves tackling high concentration fluorides and soluble poisons, which will necessitate the development of alternate matrices, advanced melters with better refractories, and better electrodes that are compatible with glasses containing fluoride. Cold crucible technology may be of use under these conditions.

With respect to fabrication of ^{232}Th/^{233}U fuel, while U, Pu fuel fabrication is well established, there is limited experience available for fuel containing ^{233}U. What makes ^{233}U always problematic is that it contains ^{232}U, which emits hard gamma radiation with a half-life of about 69 years. This means fuel fabrication has to be carried out in alpha tight hot cells with adequate shielding, unlike plutonium fuels, which can be fabricated in gloveboxes and for which an established disposal process is available. Furthermore, these facilities will generate secondary solid waste containing long lived radioisotopes with considerable gamma emission. Recovery of these long lived radioisotopes is necessary. It will be important to consider effective decontamination, minimization of waste generation, and appropriate treatment/conditioning technologies, followed by long term storage.

There is limited experience with reprocessing of thorium/thoria rods. Some are currently used in reactors for flux flattening, while some thoria rods are also employed in the blanket of the fast breeder test reactor in India. Reprocessing of thorium/thoria rods has proven very challenging. A first major problem relates to dissolution. Unlike uranium/plutonium fuels, thorium cannot be dissolved easily in boiling concentrated nitric acid; it requires the addition of fluorine. Fluorine is very corrosive, necessitating the addition of aluminium to keep the fluorine in complex form.

The unique feature of the liquid fuel used in the MSR concept is that the fertile (mainly ^{232}Th) and fissile (^{233}U/^{235}U/TRU) elements are dissolved in a fluoride based molten salt, which functions as both fuel and coolant. The presence of thorium in the fuel salt or in a blanket salt allows regeneration of ^{233}U. Therefore, it can be used as a breeder or a burner and can achieve the transition between the current U–Pu open cycle and a future Th–U closed cycle. It can even burn most of its own actinide load by the end of operation of the Th–U cycle.

The difficulties that have to be overcome before achieving an industrial grade MSR design do not necessarily represent insurmountable challenges. The present lack of explicit information on numerous technical aspects results from a lack of R&D on the subject over the past four decades. Past experiments (mainly in the USA) have shown that the molten salt technology is viable, but that the expertise in this domain has to be restored. However, there are some issues that may constitute a real handicap and their resolution could lead to decreased reactor efficiency or additional reactor cost. For example, the unknown long term reliability of the technologies involved could entail more frequent replacements of critical parts, and there are uncertainties concerning the ultimate efficiency of the fuel reprocessing in terms of the extraction of lanthanides, zirconium, alkali metals and alkaline earth metals. These issues may impact the waste assessment of the concept.

Operational waste from fuel cycle facilities

Operational wastes from the fuel cycle facilities dedicated to the various innovative reactors, combined with their fuel forms and reprocessing processes as shown in Table 9, were discussed in Section 5. In that section, the wastes were categorized as assembly hardware, hulls, noble gases, ^{3}H, iodine, ^{14}C, noble metals, ^{99}Tc, Ce/Sr and so on, as shown in Tables 9 to 20. Their routes and waste forms as well as their technology levels were studied to identify any R&D need in each of the fuel cycles. The identified R&D needs for common, aqueous processes, pyro processes and other methods are summarized in Table 26.

The fuel form and its fuel cycle process need to be selected with a view to decreasing the overall risks of radionuclides in the wastes. For example, enriched ^{15}N should be used in a nitride fuel cycle to prevent an increase in the amount of ^{14}C generated from transmutation of minor actinides.

TABLE 24. SUMMARY OF POTENTIAL INNOVATIVE REACTOR MATERIALS

System	Fuel materials						Structural materials						
	Oxide	Metal	Nitride	Carbide	Oxycarbide	Fluoride	Ferritic–martensitic SS alloys	Austenitic SS alloys	Oxide dispersion strengthened alloys (ODS)	Ni based alloys	Graphite	Refractory alloys	Ceramics
GCFR			S	P			P	P	P	P		P	P
SFR	P	P		S			P	P	P				
LFR	P	S	P				P	P	S			S	S
SCWR	P						P	P	S	S			
AHWR	P						P	P		S			
VHTR	P				P		S			P	P	S	P
MSFR						P				P		S	S
Material is new compared to current reactors			✓	✓	✓	✓						✓	✓

Notes: AHWR — advanced heavy water reactor; GCFR — gas cooled fast reactor; LFR — lead cooled fast reactor; MSFR — molten salt fast reactor; SCWR — supercritical water cooled reactor; SFR — sodium cooled fast reactor; SS — stainless steel; VHTR — very high temperature reactor.
P — Primary option; S — Secondary option.

Source: Adapted from Ref. [1].

TABLE 25. SUMMARY OF PROBLEMATIC WASTE STREAMS FROM INNOVATIVE REACTOR TYPES

Reactor type	Waste stream	Potential problem	Comments
SFR	Sodium wetted components	Reactive nature of sodium in air and water	Several sodium cooled reactors are under decommissioning and three of them were completed (including sodium cooled moderate reactors). Special processes have been developed to manage sodium contaminated materials which are treated as waste. See, for example, IAEA-TECDOC-1534.
GCFR	—	—	—
LFR	Lead wetted components	Toxic nature of lead vapours and aerosols	Simple processes already developed to manage lead (and bismuth) wetted materials which are treated as waste.[a] Standard provisions, as set forth by conventional lead industry, to manage lead vapours/aerosols.
SCWR	Containment components	Problems are known from conventional BWRs	Other challenges than those known from conventional BWRs are not expected.
AHWR	—	—	—
VHTR	Spent fuel	Large volume for final disposal	TRISO fuel is stable for final disposal, less attractive for reprocessing, but with large volume.
MSR	Salt contaminated components and fluoride salt containing actinides	Nuclear fuel material is dissolved in molten salt and should be extracted to avoid a large fluoride stream in HLW	A strong research effort is to be made on pyro processing of fluoride salts containing nuclear materials. This should include fluoride salts contaminated by actinides at low concentration.

Note: —: data not available; AHWR — advanced heavy water reactor; BWR — boiling water reactor; GCFR — gas cooled fast reactor; HLW — high level waste; LFR — lead cooled fast reactor; MSR — molten salt reactor; SCWR — supercritical water cooled reactor; SFR — sodium cooled fast reactor; TRISO — tristructural-isotopic; VHTR — very high temperature reactor.

[a] Cleaning Methods for LFR/SFR Components, ENEA Report NNFISS, LP3 – 021 2/9/2011 (public, in Italian).

TABLE 26. SUMMARY OF DEVELOPMENT NEEDS FOR FUEL CYCLE WASTES

Fuel cycle	Potential development need
Common	The conditioning of highly activated fuel structure material such as wrapper tubes and hulls should be improved to avoid hydrogen generation in the repository. The conditioning of I and Tc should be improved to decrease their long term release.
Aqueous	Vitrification should be improved to handle the increased concentration of NM. The solvent as well as its recycling technology should be developed to decrease the amount of solvent waste from MA recovery.
Pyro	Distribution of ^{129}I, ^{3}H and ^{14}C in the process should be confirmed to confine them in their waste form. Treatment of ^{129}I in the chlorine gas or in the chloride salt should be developed to confine them in their waste form. Conditioning of the salt waste should be improved to decrease the waste volume and its long term stability should be verified.

Notes: MA — minor actinide; NM — noble metal.

Problems specific to the carbide fuel cycle: In the case of aqueous processing, the combustion step prior to the nitric acid dissolution should be developed to decrease the amount of carbonate waste contaminated with ^{14}C.

Problems specific to the nitride fuel cycle: Enrichment and recycling of ^{15}N is needed to avoid the production of ^{14}C; recycling of ^{15}N in the off-gas should be developed; in the aqueous process, the combustion step prior to the nitric acid dissolution should be developed to decrease the amount of waste.

Problems specific to the VHTR and GCFR fuel cycle: Conditioning of the waste from graphite and ceramic fuel structure should be developed.

Problems specific to the MSFR fuel cycle: Extraction of HLW from fluoride salt to avoid vitrification of large amounts of fluorides.

Problems concerning ^{3}H: It is known that ^{3}H has diffused away from the ceramic fuel to sodium coolant during irradiation. But its behaviour is not certain for the metallic fuel or fuels for VHTRs and gas fast reactors (GFRs) surrounded by SiC coating or SiC tubing.

Many of the waste form technologies discussed in Section 5 are in the ‘conceptual stage.’ These should be developed to at least a ‘laboratory stage’ before conclusions can be made regarding the viability of the selected fuel cycle. Quantitative evaluation of wastes generated should confirm development objectives and adequately identify overall risks. Off-gas capture and volatile radionuclide immobilization need to be fully addressed as well.

Decommissioning waste

Lessons learned, design features and decommissioning challenges related to current facilities undergoing decommissioning should be collected and reviewed in order to facilitate design, operation, configuration management and record keeping of future innovative reactors and nuclear fuel cycle facilities. For lead or lead–bismuth cooled fast reactors, the toxicity of lead imposes the necessity to adopt dedicated procedures for processing wetted components extracted from the reactor, as well as the bulk coolant. Simple procedures have been successfully adopted in European lead laboratories; nevertheless, further development of these techniques is required to reach industrial readiness. For bulk lead, control of the temperature and of potential aerosols might be sufficient to manage the risk of dispersing toxic lead into the environment. For instance, solidifying the lead and confining it might be sufficient provision until the coolant is reused in another reactor. Finally, despite the retention capability of the coolant, the abundance of polonium in lead–bismuth might impose the need to develop and assess specific procedures for managing such a radiotoxic volatile.

7. CONCLUSIONS

Innovative reactor concepts have different objectives which should be sustainable from a resource perspective: from improving current thermal concepts operating in a once through fuel cycle to the introduction of new types of fast reactors operating in a closed fuel cycle. Innovative reactor technologies and fuel cycles are generally considered because they should have advantages over current reactor systems in terms of safety, economics, thermal efficiency, use of natural resources and waste minimization. However, the individual advantages and disadvantages of each point will differ between potential nuclear energy systems and must be considered along with all of the technical, policy and other issues. This report focuses only on the waste management aspects of innovative reactors and fuel cycles.

The key issue for the growth and sustainability of nuclear fuel cycles is the ultimate disposal of radioactive waste in a geological repository. Public acceptance can be facilitated by developing and implementing technologies that could ensure near-quantitative recovery of actinides and long lived fission products from the HLW, such that the waste monitoring period is reduced to a few hundred years. Also, effective recovery of the actinides and long lived fission products, accompanied by specific management of heat contributors by storing for decay heat, would permit a significant reduction in the requirement of repository space, which could otherwise be a limiting factor for the growth of reactor systems.

Two main pathways for waste minimization have been considered: multiple recycling of uranium/plutonium and minor actinides in fast reactors, and the use of thorium based fuel in thermal reactors to increase utilization of natural resources and minimize waste streams containing minor actinides. Both pathways will reduce the radiotoxicity of waste products, and generally reduce the quantities of long lived wastes and their resulting heat loads on repository systems. The reduction of chemical toxicity is another major focus to be considered and addressed.

The deployment of innovative reactors and associated fuel cycle facilities will create waste streams that are different (in their chemical properties, activity and volumes) from the waste created by current reactors and fuel cycle facilities. To date, considerations related to waste have focused primarily on the impact of this development on HLW associated with innovative fuel cycle concepts. The proposed approach taken in this publication also takes into consideration L&IL waste that will be generated during routine and abnormal operating conditions, as well as waste streams from decommissioning of reactors and fuel cycle facilities. Although designs of INSs are still in the conceptual stage, the systematic approach proposed serves as the first step to collect available information and develop qualitative inventory.

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Annex I

CONVENTIONAL NUCLEAR FUEL CYCLE

Radioactive waste arises from all stages of the nuclear fuel cycle. The different stages are:

— Mining and milling;
— Refining;
— Uranium conversion;
— Uranium enrichment;
— Fuel fabrication;
— Reactor operation;
— Management of spent fuel (conditioning for direct disposal or fuel reprocessing);
— Operation of waste management facilities;
— Decommissioning of nuclear fuel cycle facilities.

In the following sections, waste from each stream will be explored.

I–1. MINING AND MILLING

Mining of uranium ore is commonly carried out by either underground or open pit techniques. The amount of waste material generated is larger for open pit methods than for underground mining. A third method — in situ leaching (ISL) technology — has a very low environmental impact because no ore is brought to the surface during mining. However, its share of worldwide uranium production is still limited to about 13%, because it requires special conditions such as suitable sandstone type deposits [I–1].

The mining of ores containing from 0.1 to 0.2% uranium, on average, leaves large quantities of waste rock residue needing planned disposal. After mining, the ore is transported to a mill for chemical processing and recovery of the uranium it contains. Tailings need safe management as they contain long lived uranium and its decay products — some of which (especially radium) are toxic.

Mining and milling waste is characterized by volume that is inversely proportional to the concentration of uranium in the mined ore. At present, the uranium industry mines ores with uranium concentration as low as 0.1–0.2%. This essentially means that the volume of waste produced is equal to that of the ores treated. Taking into account the current practice of using uranium, the volume of mining and milling waste varies between 40 000 and 60 000 m^3 per 1 GW(e)a. This excludes the large quantities of waste rock with suspect radioactivity.

Table I–1 gives some parameters on the volume and radioactivity of mill tailings for various fuel cycles.

TABLE I–1. SOME PARAMETERS OF MILL TAILINGS FOR VARIOUS FUEL CYCLES, NORMALIZED TO 1 GW(e)

Parameters	LWR		HWR (CANDU)		FBR
	Once through	U–Pu cycle	Once through	U–Pu cycle	U–Pu cycle
Uranium extracted from ore (t)	205	120	180	75	1.2
Waste mill tailings (m^3)	58 000	34 000	51 000	21 000	340
Activity of ^{230}Th and ^{226}Ra, each (GBq)					
Total activity: beta, alpha (GBq)					

Note: LWR — light water reactor; HWR — heavy water reactor; FBR — fast breeder reactor.

I–2. REFINING

Refining is defined as the processing of uranium ore concentrates to produce uranium trioxide (UO_3) or uranium dioxide (UO_2). This process may be carried out on a single site or as part of an integrated process involving different sites. A general sequence of different processes resulting in UO_3 and UO_2 production considers:

- Purification.
- UO_3 and UO_2 production by:
  - Thermal denitration (TDN) process;
  - Ammonium diuranate (ADU) process;
  - Ammonium uranyl carbonates (AUC) process.

The typical waste arising from the refining processes is given in Table I–2 [I–2], for a hypothetical facility that refines 1000 t of uranium. A major part of waste generated during the refining process is associated with the purification stage, or with formation of uranyl nitrate liquor (UNL). After the purification stage, UNL is quite pure and very low amounts of waste are linked with the production either of UO_3 or of UO_2.

I–3. URANIUM CONVERSION

Although uranium ore concentrate (yellow cake) is fairly pure, it requires further purification to reach the very high standards required for nuclear fuel. This is achieved by dissolving the yellow cake in nitric acid, filtering, and treating the solution with chemical solvents. The product is the compound uranyl nitrate, which is usually more than 99.95% pure. The uranyl nitrate is reconverted to uranium oxide (UOX) and that, in turn, is converted to readily volatile uranium hexafluoride which is used in the enrichment process [I–3]. This UF_6 is the only uranium compound that is suitable for performing enrichment, because of its thermal stability and relatively high volatility. All current enrichment processes are based on the use of uranium hexafluoride. If enrichment is not required, uranium dioxide may be produced from the uranyl nitrate and shipped directly to a fuel element fabrication plant.

The production process has the following stages:

- Reduction of UO_3 to UO_2;
- Conversion of UO_2 to UF_4;
- Fluorination of UF_4 to UF_6.

The typical waste arising from the conversion processes is given in Table I–3.

TABLE I–2. TYPICAL WASTE ARISINGS FROM THE REFINING PROCESSES (FOR 1000 t U)

Waste arising	Quantity
Drums	70 t
Insolubles + filter aid	50 t
Liquid effluents	3000 to 10 000 m³
Sludge	300 t
Liquid nitrates	200 t

TABLE I–3. TYPICAL WASTE ARISING FROM THE CONVERSION PROCESS (FOR 1000 t U)

Waste arising	Quantity (t)
Solid CaF_2	10
Sludge CaF_2, $Ca(OH)_2$, H_2O with small amounts of U	20 to 50
Sludge CaF_2, $Ca(OH)_2$, H_2O without U	30

I–4. URANIUM ENRICHMENT

Enrichment involves increasing the proportion of ^{235}U, from the natural level of 0.7% to an average level of 3 to 5%, in UF_6. This can be done using one of two different industrial methods: gaseous diffusion or centrifugation.

Gaseous diffusion enrichment is based on the different diffusion rates of gaseous $^{235}UF_6$ and $^{238}UF_6$ through membranes. In the centrifuge process, enrichment is achieved by differential centrifugation [I–4]. After separation, the portion enriched in the ^{235}U isotope is transferred to the fuel element fabrication plant, and the larger part (enrichment tails) is stored or processed. Uranium metal is produced by the reaction of the UF_4 with an alkali earth metal, using a thermal type process. In addition to uranium metal, this produces uranium contaminated slag of the alkali metal fluoride. One of two processes can produce UO_2: the AUC process, and the integrated dry route process whereby UF_6 is ‘burned’ in steam under a reducing hydrogen atmosphere. Both processes give rise to hydrogen fluoride as a by-product [I–4].

Centrifuge and gaseous diffusion processes produce only very minor quantities of waste [I–2]. This is because these plants handle a single process medium (UF_6) which is completely contained in a high integrity system throughout the operation. Since the processes are physical, not chemical, there are no auxiliary inflows of material or any rejects of intermediate or waste products in the accepted sense. The minor quantities of waste which do arise result from the light gas which passes through a small scrubbing system to ensure that only clean exhaust is released into the atmosphere. Very small quantities of uranium (^{234}U, ^{235}U, ^{238}U) are vented to the atmosphere from gaseous diffusion plants. The radioactive discharges from centrifuge enrichment facilities are even smaller.

Enrichment of 1000 t of uranium in the form of UF_6 leads to the generation of around 850 t of depleted uranium with a ^{235}U content of approximately 0.2%. This material may be classified as a by-product or as waste. Laser technology — now under development on a laboratory or a pilot scale — potentially provides an opportunity to reduce the content of ^{235}U in depleted uranium by a factor of at least five, and to exclude highly toxic fluoride from the enrichment process.

I–5. FUEL FABRICATION

Fuel fabrication produces a fuel for loading into a nuclear reactor. Uranium dioxide and metallic uranium are two products that are commonly used as starting materials for fuel fabrication. Natural uranium is only used for the production of metallic uranium fuel. When uranium dioxide is used, it can be either natural or enriched. Some light water reactors (LWRs) and fast reactors use mixed uranium–plutonium oxide (MOX) fuel. Plutonium from reprocessing is recycled into MOX fuel in Belgium, France, Germany, Japan, the Russian Federation and Switzerland. There is also broad experience in operating reactors with MOX fuel.

The generic oxide and mixed oxide fuel fabrication process consists of the following steps:

- Preparing the material suitable for pellet formation (correct morphology, blend, purity, etc.). This is achieved, for example, by granulation and grinding, with blending of uranium and plutonium oxide powders, where necessary (MOX fuels).
- Compacting to form a pellet either with or without a binder to sustain the integrity of the pellet.
- Sintering followed by grinding to produce pellets of the required dimensions
- Assembling pellets in pins (Zircaloy or stainless steel tubes) with appropriate spacers and springs. These pins are assembled into fuel elements using bracing, end fittings and caps appropriate to the fuel [I–4].

TABLE I–4. TYPICAL WASTE ARISING FROM THE FUEL FABRICATION FOR 1000 t U THROUGHPUT

Waste arising	Quantity
Extraction residues	10 m^3
Sludge	1 m^3
Graphite and/or activated charcoal	300 t
Zircaloy	1 t
Stainless steel	1 t
Miscellaneous metal scrap	40 t
Ventilation filters	100 to 200 m^3
Mixed combustible material	300 m^3

The fuel fabrication stage has the potential to produce a significant quantity of material scrap. Most of this scrap is not considered waste because it has significant value, and because the majority of materials can be recycled within the process. Low and intermediate level waste (L&ILW) from fuel fabrication includes filter media from wash water cleanup, waste oils, spent acids and bases, spent analytical solutions, decontamination and cleaning solutions and discarded scrap metals and equipment. Any of these wastes may be contaminated with hazardous chemicals and uranium. Plutonium contamination is present from facilities manufacturing mixed oxide fuel.

Table I–4 lists typical waste arising at a fuel fabrication facility per 1000 t of uranium throughput [I–5].

I–6. SPENT FUEL MANAGEMENT

Spent fuel management includes the technical operations beginning with the discharge of spent fuel from a power reactor and ending with either the reprocessing of spent fuel and recycling of plutonium and uranium in new MOX fuel (closed cycle) or the direct disposal of the spent fuel elements (open, once through cycle). A closed cycle also requires a solution for the final disposal of the (high level) waste resulting from reprocessing (i.e. the fission products and fissile material that are no longer recycled).

Spent fuel reprocessing

At present, all commercial reprocessing plants use the aqueous plutonium uranium redox extraction (PUREX). In this process, uranium, plutonium and sometimes neptunium, if needed, are separately extracted from the solution resulting from the dissolving of the fuel assemblies in nitric acid. The solution contains more than 99% of the fission products and transuranic elements (TUEs) formed in the reactor. It is then treated by the organic solvent tri-n-butyl phosphate (TBP). The use of a PUREX process for reprocessing oxide spent fuel from fast breeder reactors is being considered in some countries; it was used for the MOX fuel from the Dounreay fast reactor in the United Kingdom.

Vitrification of high level waste

Vitrification has become the reference process for the conditioning of high level aqueous effluents from reprocessing of spent fuel. In all designs, the solid content of liquid high level waste (HLW) is eventually incorporated into a matrix of borosilicate glass at a temperature near 1100°C (the Russian Federation uses phosphate glass). The vitrification may be attained in a one step process, in which the effluent is directly injected into the molten borosilicate bath. It may also be achieved in two steps, the first one consisting of evaporation and calcinations and the second one of the incorporation of the calcined residue in the borosilicate melt.

In most existing plants, vitrification is carried out in melters based on induction or Joule heating. The susceptor and the process pot are made of a high Ni–Cr alloy (Inconel-690) so as to withstand high temperature as well as oxidizing and corrosive conditions. The calcined mass is fused into glass at about 1223 K, and is soaked at 1223–1273 K for about eight hours to achieve homogenization. The molten mass is drained into stainless steel canisters by operating a freeze valve. The canister filled with glass is allowed to cool slowly in an insulated assembly. This is welded remotely by means of the pulse TIG method. Several such canisters are further enclosed in secondary steel containers called overpacks. An elaborate off-gas cleaning system consisting of a condenser, scrubber, chiller, demister and absolute HEPA filter is used to treat the gases before discharge through a tall stack.

Typically, a 1000 MW(e) LWR uses about 27 to 30 t of fuel per year. The volume of liquid HLW from reprocessing this fuel, including compacted hulls, is about 15 m^3. The HLW is typically vitrified; vitrification reduces the volume of HLW to about 3 m^3. For liquid HLW that has been in storage for 10 years, the specific activity will be on the order of 7×10^{16} $Bq{\cdot}m^{-3}$ and in the vitrified product about 4×10^{17} Bq/t.

Solid waste processing

During reprocessing, other less active waste is generated (e.g. empty hulls, spacers, insolubles) which nevertheless contains appreciable amounts of long lived radionuclides (fission products, activation products and some residual fissile materials). Empty hulls contain beta, gamma and alpha activities. Activated and contaminated metals derived from cladding of SNF generally have low or moderate radiotoxicity, since metallic materials are either of a low radiotoxicity, such as magnesium and iron, or of a low solubility, such as zirconium and stainless steels. Liquid L&ILW is also produced. However, some materials, such as ^{60}Co and Zr isotopes produce hard gammas that result in radiation protection issues and potential implications for waste handling.

The hulls and cladding of the fuel elements need to be separated from the fuel prior to dissolution of the fuel in nitric acid. To separate zirconium cladding from the oxide fuel, three methods are used or under development:

(a) Mechanical fragmentation followed by dissolution in nitric acid and separation of liquid and solid phases;
(b) Embrittlement of zirconium cladding by gas (oxygen or nitrogen), crushing followed by dissolution of the crushed pieces in nitric acid and separation of liquid and solid phases;
(c) Dissolution of zirconium cladding in metallic melts (e.g. zinc) followed by separation of pellets from melted metal.

To separate stainless steel cladding from oxide fuel, three methods are applied:

(a) Fragmentation followed by the same procedures as above;
(b) Melting of stainless steel followed by separation of pellets from the metallic melt;
(c) Dissolution of stainless steel cladding in metallic melts (e.g. zinc) followed by separation of pellets from melted metal.

Liquid waste processing

Nitrate is a significant component of a number of reprocessing waste streams, because nitric acid is used for spent fuel dissolution. Spent solvents can arise from the solvent extraction processes used for chemical separations. The most commonly used extraction solvent is TBP, which is diluted for the extraction process, usually with a lightly saturated hydrocarbon such as dodecane or a mixture of paraffin hydrocarbons.

Treatment of liquid radioactive waste involves the following steps:

— Radionuclide concentration by evaporation, ion exchange, sorption, etc.;
— Solidification of concentrates by evaporation, bituminization, cementation and inclusion (in e.g. polymers, glass, ceramics, synthetic rocks and glass–metal matrices);
— Partial return of the purified water and other substances for reuse.

In several cases, it is considered useful to perform waste partitioning (i.e. extraction of one or several groups of radionuclides for subsequent separate treatment). Liquid waste with a low salt concentration is treated by a combination of co-deposition, filtration, ion exchange, sorption and other methods, with the final aim of producing water for recycling and or release to an authorized discharge point, and salt concentrate to be processed for further solidification (in the form of sludge, pulps, etc.). Selecting a solidification method for liquid waste depends on the radionuclide content and chemical composition, activity and waste volume.

A variety of L&ILW liquid waste is suitable for incorporation into cement matrices. There is widespread potential for using this process, and the concreted waste form has certain inherent qualities (e.g. resistance towards radiation, compatibility with many types of environmental conditions). Special chemical resistant cements are now also being used (e.g. super sulphate cement and slag cement) for conditioning waste contaminated with transuranium elements. The acceptability of the concrete option depends upon the characteristics of the intended disposal environment and compatibility with those characteristics.

Bituminization is applied for conditioning chemical precipitates and evaporator concentrates. The conditioned waste form has a very low permeability and solubility in water, and is compatible with most environmental conditions. Some restrictions must, however, be exercised with regard to the incorporation of strongly oxidizing components (e.g. nitrates and salts). Furthermore, questions may be raised concerning the very long term physicochemical and radiation stability of bitumen, as it might not remain an efficient protective barrier for radionuclides with very long half-lives, such as ^{239}Pu. Bituminization could be considered for the immobilization of very low heat producing concentrates. Its use should be restricted preferably to materials with low alpha contamination. At the same time, the bituminization process reduces the volume of waste concentrates which also reduces costs in the subsequent disposal steps.

Waste contaminated with a substantial amount of plutonium is sometimes considered a particular problem. For the recovery of plutonium from the waste streams, two processes have been tried:

(a) Acid (sulphuric and nitric acid) digestion followed by recovery of the fissile materials;
(b) Low temperature incineration of the waste, followed by leaching of the ash to recover the fissile material.

Disposal

All categories of radioactive waste, from L&ILW to HLW, are generated as a result of spent fuel reprocessing. Long lived radionuclides are contained in these wastes. After conditioning, HLW and long lived L&ILW require disposal in a deep geological repository. If an open cycle is selected, the spent fuel also needs to be conditioned (encapsulated and packed) and placed in a geological repository.

The first deep repository is operating in the United States of America: the waste isolation pilot plant. The repository accepts for disposal only waste coming from the defence programme that contains TUEs. At present, no deep geological repositories exist for HLW or for spent fuel. However, several are under active development, such as in Finland and Sweden, and site selection processes are under way in several other countries, such as France and Japan. Deep repositories for L&ILW are also being developed or planned in several countries, such as Canada, Germany and the United Kingdom. These facilities would be capable of accepting long lived L&ILW

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Annex II

EXAMPLES OF INNOVATIVE NUCLEAR POWER REACTORS

There are several international initiatives and collaborative projects that are coordinating efforts on the development of innovative reactor systems. One such initiative is the Generation IV International Forum (GIF) [II–1, II–2].

The GIF identifies a number of reference technologies that could be deployed over the next several decades:

- Gas cooled fast reactor system (GCFR) (Fig. II–1);
- Lead cooled fast reactor system (LFR) (Fig. II–2);
- Molten salt fast reactor system (MSFR) (Fig. II–3);
- Sodium cooled fast reactor system (SFR) (Fig. II–4);
- Supercritical water cooled reactor system (SCWR) (Fig. II–5);
- Very high temperature reactor system (VHTR) (Fig. II–6).

Variations of these concepts appear in a number of national and international programmes. They are described below as examples of the types of innovative reactor systems that are considered in this publication. For each of these six reactors systems, reactor parameters and reference values have been defined. Those parameters which are important for waste management are provided in tables for every reactor type.

The design features of a power reactor have the greatest influence on the volumes and characteristics of waste generated during its operational lifetime and decommissioning, while processes involved in manufacturing and reprocessing nuclear fuel define the volumes and characteristics of waste from fuel cycle activities.

II–1. GAS COOLED FAST REACTOR (GCFR)

The GCFR system (Fig. II–1) features a fast neutron spectrum helium cooled reactor and closed fuel cycle for efficient management of actinides and conversion of fertile uranium. Core configurations are being considered based on pin or plate based fuel assemblies, with a total core power of 300–600 MW(e). The high outlet temperature of the helium coolant makes it possible to deliver electricity, hydrogen or process heat with a high conversion efficiency. The GCFR uses a direct cycle helium turbine for electricity and can also use process heat for the thermo-chemical production of hydrogen. Through the combination of a fast neutron spectrum and full recycling of actinides, GCFR minimizes the production of long lived radioactive waste isotopes. The GCFR's fast spectrum also makes it possible to utilize available fissile and fertile materials (including depleted uranium from enriched plants) that are two orders of magnitude more efficient than thermal spectrum gas reactors with a once through fuel cycle. The GCFR system has strong sustainability because of its closed fuel cycle and excellent performance in actinide management. It is rated well in safety, economics, proliferation resistance and physical protection. Some parameters of GCFR are given in Table II–I.

II–2. LEAD COOLED FAST REACTOR (LFR)

The LFR system (Fig. II–2) features a fast neutron spectrum and a closed fuel cycle for efficient management of actinides and conversion of fertile uranium. The system uses a lead or lead–bismuth eutectic liquid metal to cool the reactor. The reactor is cooled by either natural (for small size systems) or forced (for medium large systems) convection and sized between 50 and 1200 MW(e), with a reactor outlet coolant temperature of up to 550°C, possibly ranging up to 800°C, depending upon the success of material research and development (R&D). The LFR system has strong sustainability because a closed fuel cycle is used. For proliferation resistance and physical protection its long life core provides strong sustainability. Safety is enhanced by the choice of a relatively inert coolant.

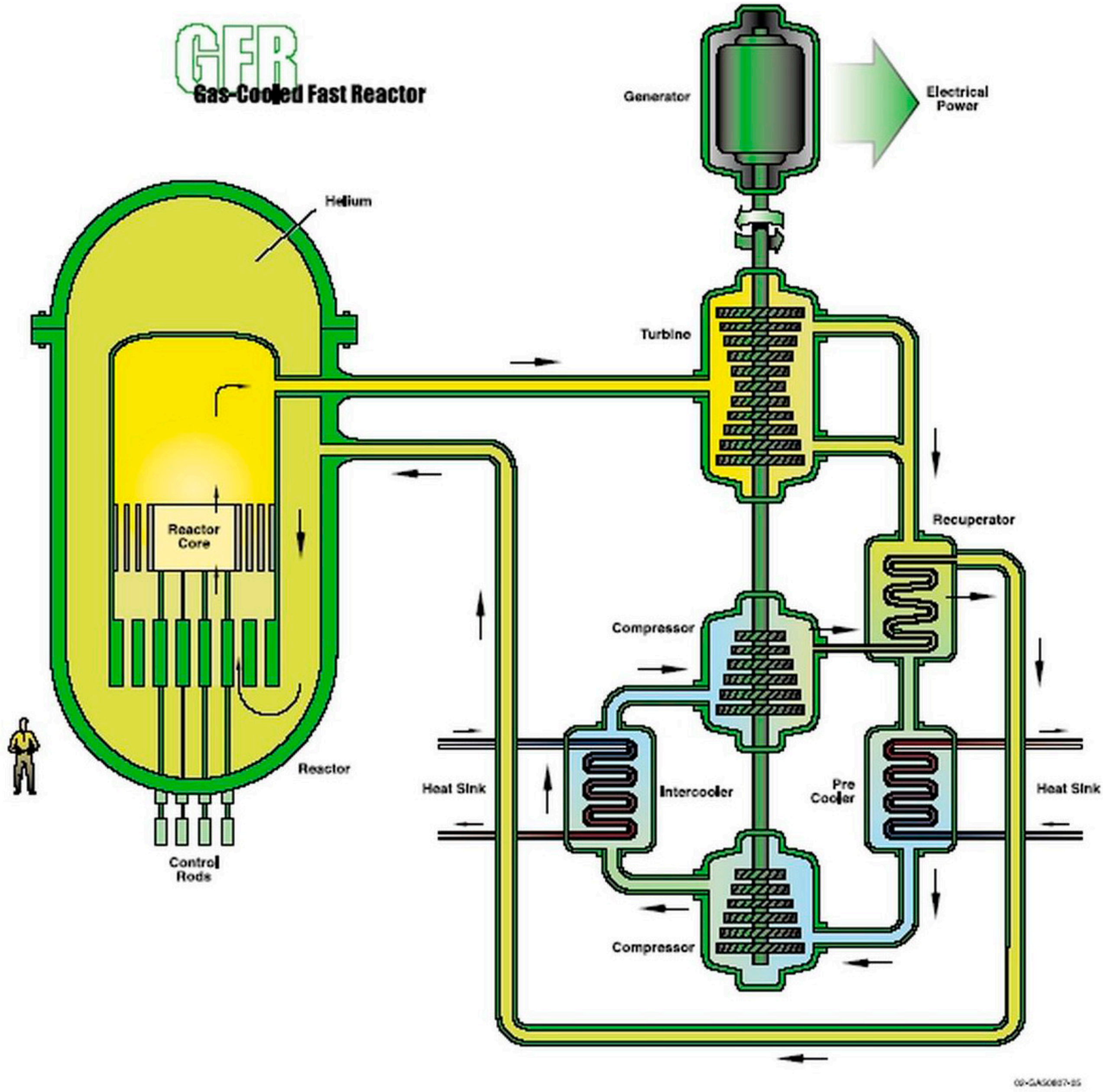


FIG. II–1. Gas cooled fast reactor system, Ref. [II–1].

It is primarily envisioned for applications such as electricity generation, hydrogen production and actinide management with good proliferation resistance [II–1]. Three different designs have been assumed as a reference for GIF activities: SSTAR (USA, small size reactor design), BREST OD300 (Russian Federation, medium size reactor design) and ELFR (EURATOM, large size reactor design). Some parameters of such LFR designs are given in Table II–2, together with other reactor designs presently under development.

II–3. MOLTEN SALT FAST REACTOR (MSRF)

The main characteristic of the MSFR system (Fig. II–3) is its molten fuel. In the MSFR, liquid fluoride fuel flows through a central compact canal where heat is produced and then directed through heat exchangers. The fuel flows freely from the bottom to the top of the central component without a solid moderator. The circulation of the salt is divided into 16 groups of pumps and heat exchangers located around the core. The fuel salt completes a full cycle

TABLE II–1. GAS COOLED FAST REACTOR: REACTOR PARAMETERS AND REFERENCE VALUES

Reactor parameters	Reference value
Reactor power	300–600 MW(th)
Net plant efficiency (direct cycle helium)	48%
Coolant inlet/outlet temperature and pressure	490°C/850°C at 90 bar
Average power density	100 MW(th)/m^3
Fuel type	Carbide/nitride
Reference fuel compound	UPuC/SiC (70/30%) with about 20% Pu content
Cladding	—
Volume fraction, fuel/gas/SiC	50/40/10%
Conversion ratio	Self-sufficient
Burnup, damage	50 MW·d/t HM; 60 dpa
Fuel processing	Pyro/aqueous

Note: —: data not available.

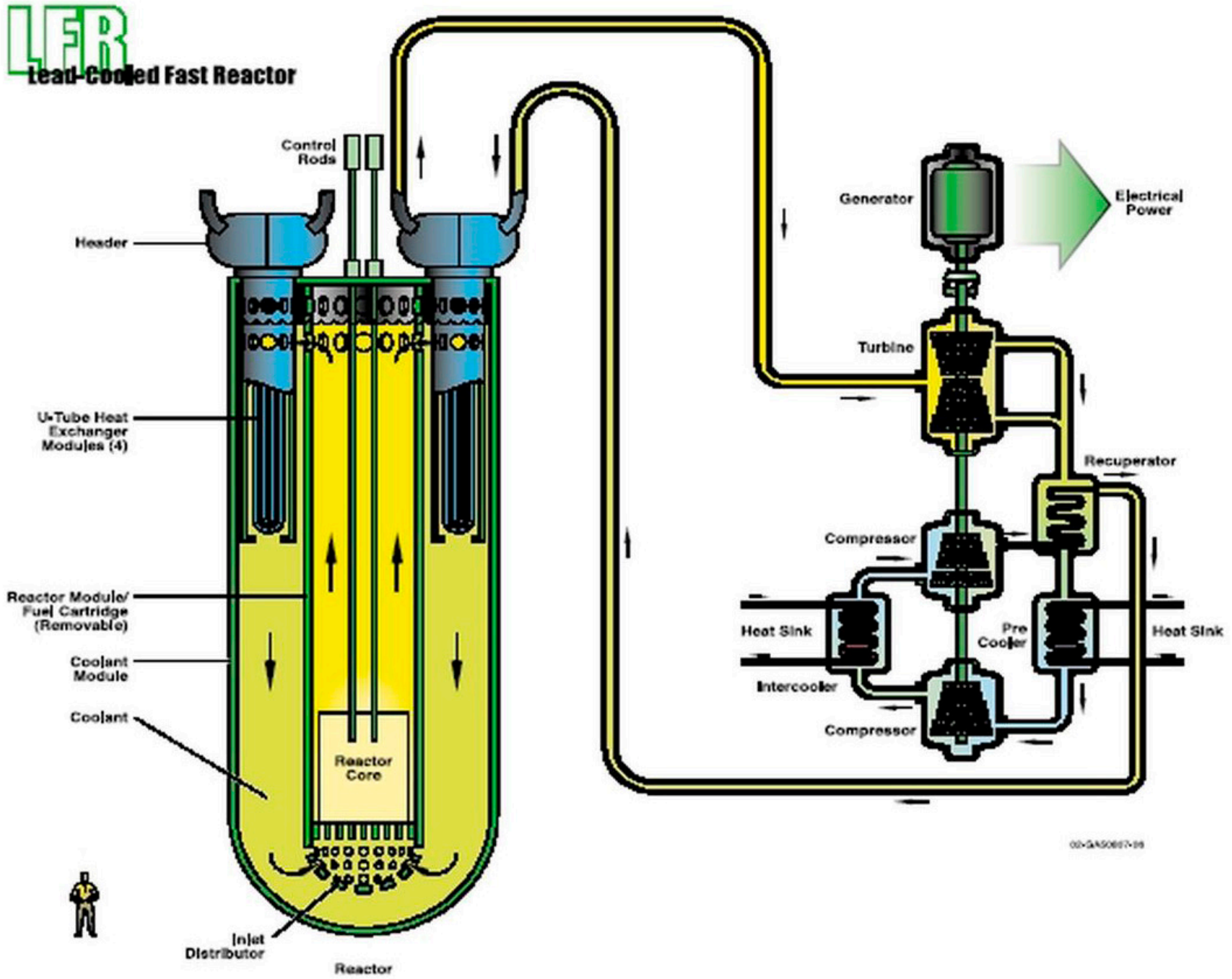


FIG. II–2. Lead cooled fast reactor system, Ref. [II–1].

TABLE II–2. LEAD COOLED FAST REACTORS: REACTOR PARAMETERS AND REFERENCE VALUES

Reactor parameters	Reference values					
	GIF reference systems				Other systems	
	SSTAR	BREST-300	ELFR	SVBR-100	ALFRED	G4M
Rating (MW(th))	45	700	1500	280	300	70
Coolant	Pb	Pb	Pb	Pb–Bi	Pb	Pb–Bi
Pressure (bar)	1	1	1	1	1	1
Outlet temp. (°C)	564	540	480	490	480	500
Fuel	Nitride	Nitride	MOX/MOX + MA	UO_2/MOX	MOX	Nitride
Cladding	HT-9	Cr_{12}MoVNbB	T91 (15–15/Ti)	HT-9	15–15/Ti	HT-9
Avg./max. burnup (GW·d/t HM)	81/131	53/84	52/90	69/114	73.3/103	—
Conversion ratio	—	~1	1.07	0.84/>1	0.6	—
Lattice	Open	Closed	Closed	Open	Closed	—
Primary flow	Natural	Forced	Forced	Forced	Forced	Forced
Max./avg. linear power (W/cm)	—	380/n.a.	257/150	n.a./260	350/214	—
Fuel reprocessing	Pyro	Pyro	Wet	Wet	Wet	Pyro

Note: MA — minor actinide; MOX — mixed oxide; n.a.: data not applicable; —: data not available.

in three to four seconds. At any time, half of the total fuel salt volume is in the core and half in the external fuel circuit (salt collectors, salt bubble separators, fuel heat exchangers, pumps and pipes) (Table II–3). There is a fertile blanket all around the central canal. The MSFR fuel salt is composed of fluorides of ^{7}Li and heavy nuclei (HN), the heavy nuclei being fertile thorium and fissile material, ^{233}U, enriched uranium and/or plutonium, and minor actinides. The (HN)F_4 proportion is set at 22.5 mole% (eutectic point), corresponding to a melting temperature of 565°C.

One advantage of a liquid fuel is that its configuration can be modified by draining the fuel salt by gravity into tanks located below the reactor. Two fuel configurations are thus available:

(1) The critical fuel configuration for heat production;
(2) The subcritical fuel configuration in the draining tanks, designed to optimize heat and neutron evacuation and allow passive and active heat extraction [II–3].

In the critical configuration, power is extracted by an intermediate coolant (a fluoride salt), which allows a connection with a conventional conversion system.

The present reference concept is a 1300 MW(e) reactor, with the reprocessing flowsheet proposed in Section 4.8.2 of this publication. The calculations are made for a fuel containing only ^{233}U, without transuranic (TRU). Starting the reactor with plutonium and minor actinides is possible; the salt composition evolves slowly with time [II–1]. Some parameters of the MSFR are given in Table II–4. Some parameters of molten salt reactors (MSRs) of 1000 MW are given in Table II–4.

II–4. SODIUM COOLED FAST REACTOR (SFR)

The SFR system (Fig. II–4) features a fast neutron spectrum and a closed fuel cycle for efficient management of actinides and conversion of fertile uranium. A full actinide recycling fuel cycle is envisioned with two major options. One is an intermediate size (150 to 500 MW(e)) sodium cooled reactor with uranium–plutonium minor actinide zirconium metal alloy fuel, supported by a fuel cycle based on pyrometallurgical processing. The second is a medium to large (500 to 1500 MW(e)) sodium cooled fast reactor with mixed uranium–plutonium oxide fuel, supported by a fuel cycle based upon advanced aqueous processing at a central location serving a number

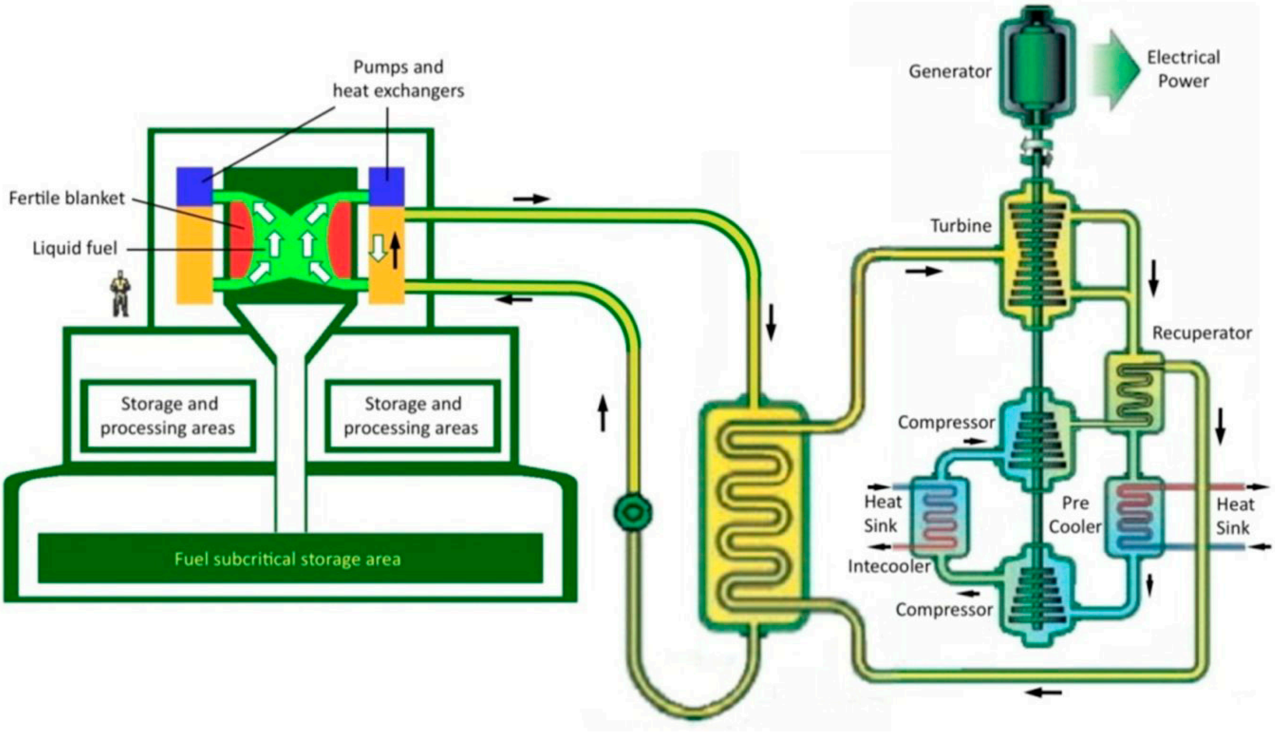


FIG. II–3. Molten salt reactor system.

TABLE II–3. MOLTEN SALT REACTORS: REACTOR PARAMETERS AND REFERENCE VALUES

Reactor parameter	Reference value
Net power	1000 MW(e)
Power density	22 MW(th)/m^3
Net thermal efficiency	44 to 50%
Fuel type	Molten salt (fluorides)
Cladding	n.a.
Fuel salt	—
Inlet temperature	565°C
Outlet temperature	700°C (850°C for hydrogen production)
Vapour pressure	<0.1 psi
Moderator	Graphite
Power cycle	Multi-reheat, recuperative helium, Brayton cycle
Neutron spectrum burner	Thermal–actinide
Fuel reprocessing	Pyro

Note: n.a.: data not applicable, — : data not available.

TABLE II–4. MOLTEN SALT FAST REACTOR: REACTOR PARAMETERS AND REFERENCE VALUES

Reactor parameter	Reference value
Net power	1300 MW(e)
Mean power density in the central canal	330 MW(th)/m^3
Net thermal efficiency	43%
Fuel for the reference reactor	77.5 ^{7}LiF – 20 ThF_4 – 2.5 $^{233}UF_4$
Intermediate fuel salt coolant	LiF–NaF–KF
Inlet temperature of the fuel salt	730°C
Outlet temperature of the fuel salt	800°C
Hydraulic height	5 bar
Structure materials	Nickel based alloys, molybdenum, SiC
Fuel reprocessing	Pyro

of reactors. The outlet temperature is 550°C for both. The SFR system has strong sustainability because of its closed fuel cycle and excellent potential for actinide management [II–1]. It is rated good in safety, economics and proliferation resistance and physical protection. It is primarily envisioned for electricity production and actinide management. Some parameters of SFRs are given in Table II–5.

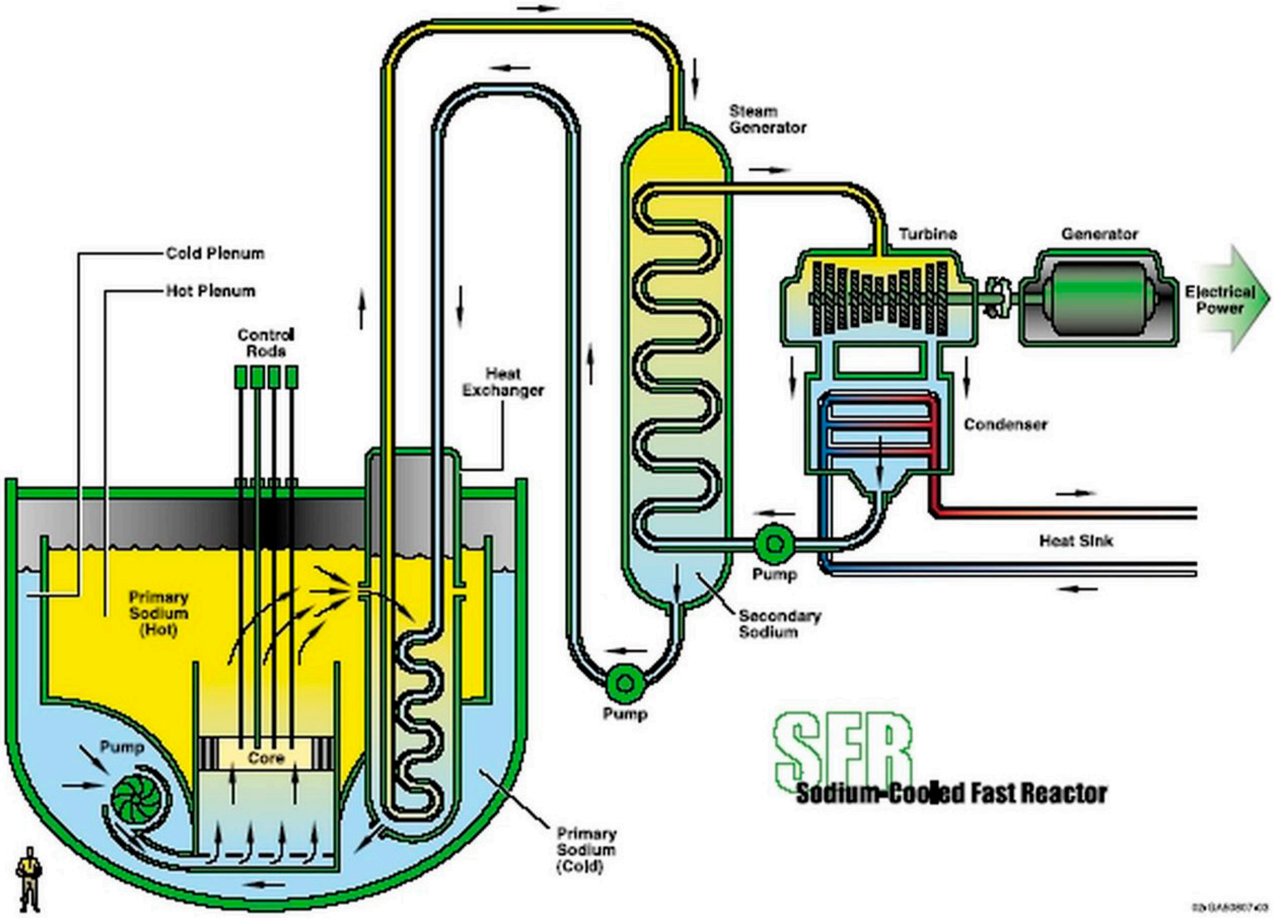


FIG. II–4. Sodium cooled fast reactor system, Ref. [II–1].

TABLE II–5. SODIUM COOLED FAST REACTOR: REACTOR PARAMETERS AND REFERENCE VALUES

Reactor parameter	Reference value
Outlet temperature	530–550°C
Pressure	~1 atmosphere
Rating	1000–5000 MW(th)
Fuel	Oxide, MOX or metal alloy
Cladding	Ferritic or oxide dispersion strengthened (ODS) ferritic
Average burnup	~150–200 GW·d/t HM
Conversion ratio	0.5–1.30
Average power density	350 MW(th)/m^3
Fuel processing	Aqueous/pyro

II–5. SUPERCRITICAL WATER COOLED REACTOR (SCWR)

The SCWR system (Fig. II–5) features an open cycle with a thermal neutron spectrum reactor as a primary option. The system uses high temperature, high pressure water for cooling the reactor, which operates above the thermodynamic critical point of water to achieve a thermal efficiency approaching 44%. The reference plant has a 1700 MW(e) power level and a reactor outlet temperature of 550°C. The SCWR system is highly ranked in economics because of high thermal efficiency and plant simplification [II–1]. Some parameters of SCWRs are given in Table II–6.

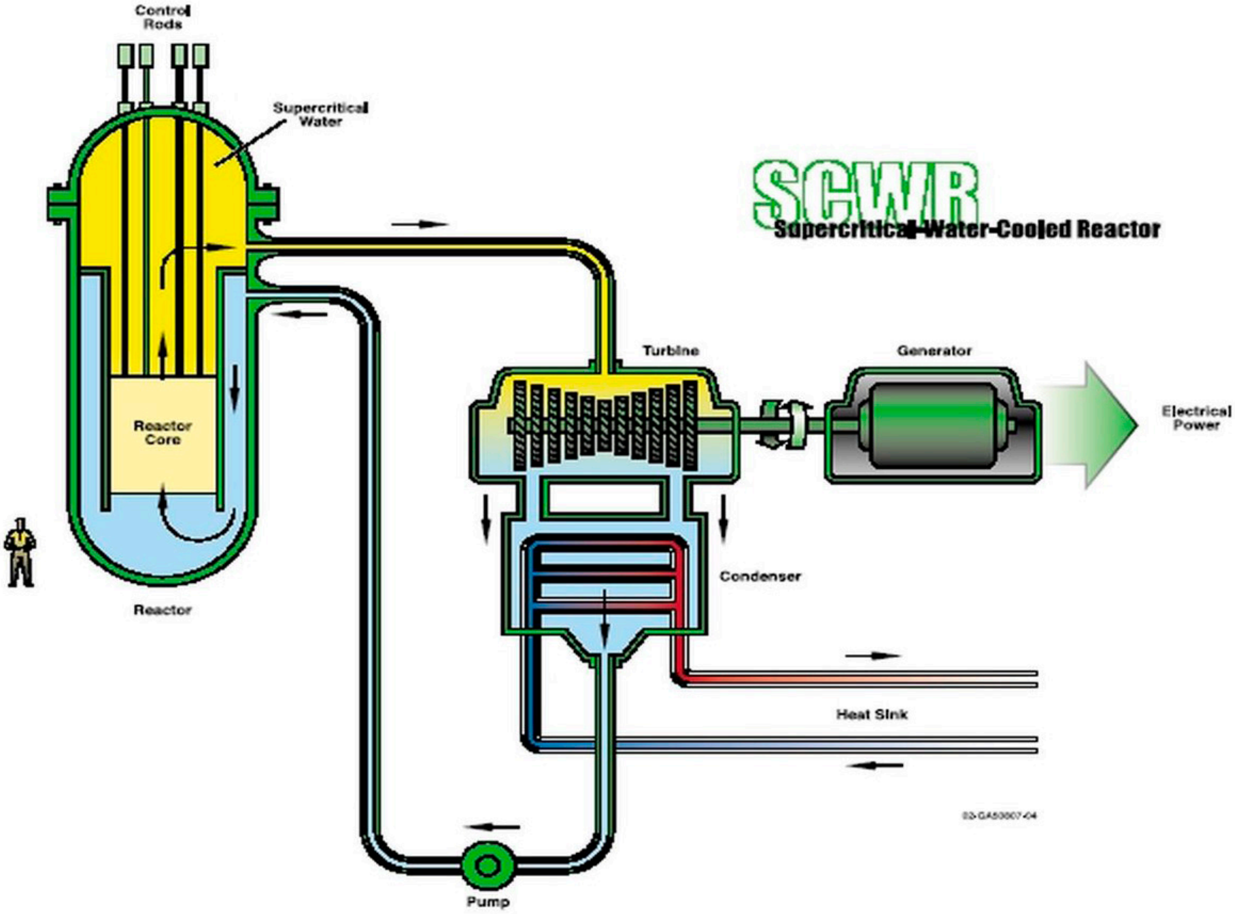


FIG. II–5. *Supercritical water cooled reactor system, Ref. [II–1].*

TABLE II–6. SUPERCRITICAL WATER COOLED REACTOR: REACTOR PARAMETERS AND REFERENCE VALUES

Reactor parameter	Reference value
Unit power and neutron spectrum	1700 MW(e), thermal spectrum
Net efficiency	44%
Coolant inlet/outlet temperatures and pressure	280°C/510°C 25 MPa
Average power density	~100 MW(th)/m^3
Fuel type	Oxide (UOX, MOX)
Average burnup	50 MW·d/T
Fuel reprocessing	Aqueous

II–6. VERY HIGH TEMPERATURE REACTOR (VHTR)

The VHTR system (Fig. II–6) uses a thermal neutron spectrum and once through uranium cycle. The VHTR system can supply nuclear heat and electricity over a range of core outlet temperatures between 700 and 950°C. The original target was for an outlet temperature above 1000°C to perform thermochemical hydrogen production at high efficiency and without carbon emission. The current market conditions did, however, show a large market for electricity production and industrial processes based on high temperature steam with more modest outlet temperatures. So, in the past few years the focus has changed from designs with higher outlet temperatures and direct gas turbines (such as the prismatic block fuel high temperature helium cooled reactor and pebble bed modular reactor) to lower outlet temperature designs such as the high temperature reactor pebble bed module under construction in China and the next generation nuclear plant in the USA. The VHTR designs are modular, with reactor powers of 200–600 MW(th). Two different fuel and core configurations are used, the pebble bed and prismatic block type. The VHTR technology is based on TRISO coated particle fuel, graphite as the core structure, helium coolant, as well as the dedicated core layout and lower power density to achieve decay heat removal in a natural way. The VHTR mostly focuses on an open low enriched uranium fuel cycle but can support alternative fuel cycles such as U–Pu, Pu, MOX and U–Th. Some parameters of single design of an older concept VHTR are given in Table II–7.

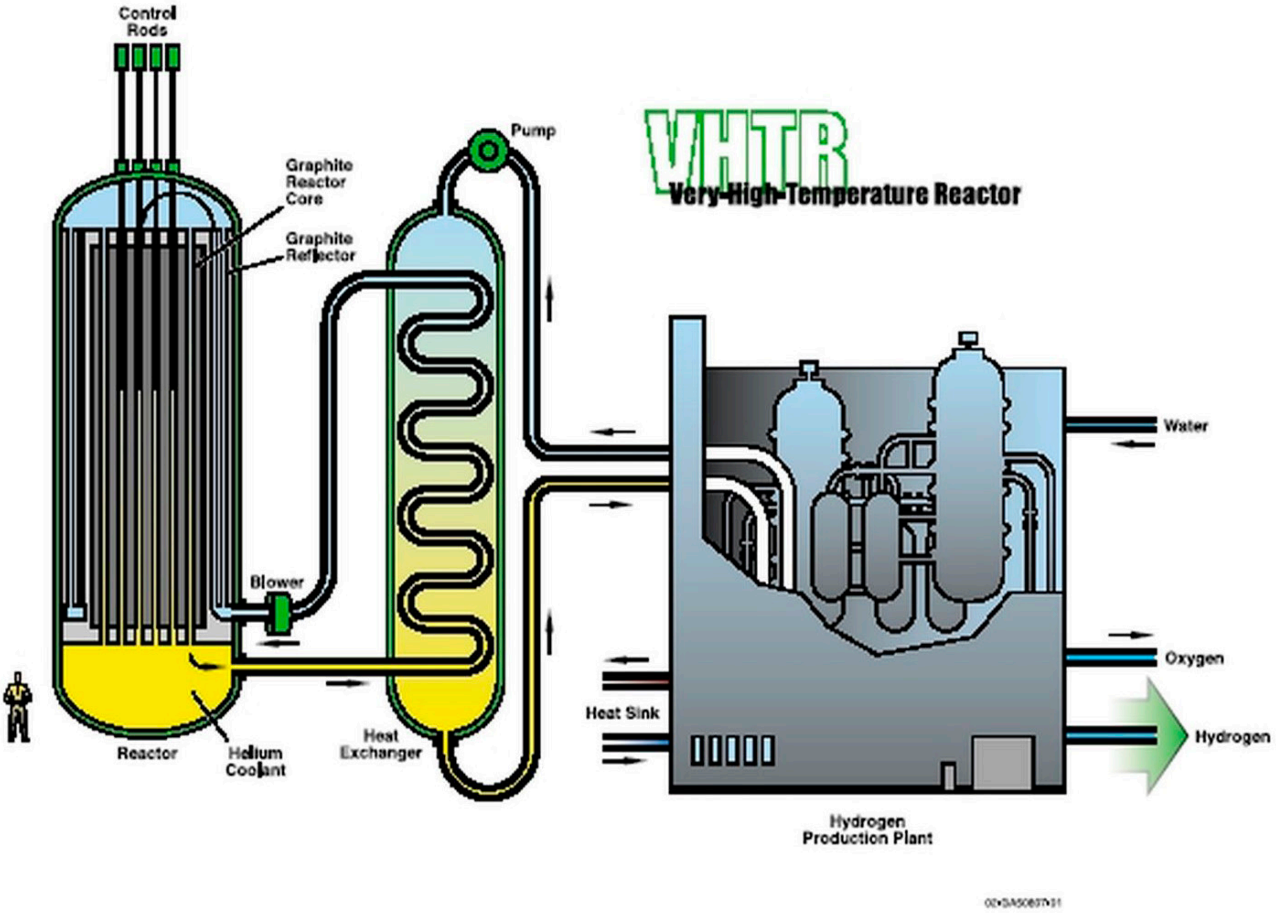


FIG. II–6. *Very high temperature reactor system, Ref. [II–1].*

TABLE II–7. VERY HIGH TEMPERATURE REACTOR: REACTOR PARAMETERS AND REFERENCE VALUES

Reactor parameter	Reference value
Reactor power	600 MW(th)
Coolant inlet/outlet temperature	640/1000°C
Core inlet/outlet pressure	Dependent on process
Helium mass flow rate	320 kg/s
Average power density	6–10 MW(th)/m^3
Fuel type	Carbide
Reference fuel compound	ZrC coated particles in blocks, pins or pebbles
Cladding	—
Average burnup	>100 MW·d/t
Net plant efficiency	>50%

Note: —: data not available.

REFERENCES TO ANNEX II

[II–1] GENERATION IV INTERNATIONAL FORUM, A Technology Roadmap for Generation IV Nuclear Energy Systems, USDOE, Office of Nuclear Energy, Washington, DC (2002).

[II–2] OECD NUCELAR ENERGY AGENCY, Technology Roadmap Update for Generation IV Nuclear Energy Systems, OECD/NEA, Paris (2014).

[II–3] HEUER, D., et al., Towards the thorium fuel cycle with molten salt fast reactors, Ann. Nucl. Energy **64** (2014) 421–429.

Annex III

EXAMPLES OF INNOVATIVE NUCLEAR FUEL CYCLES

In conventional thermal reactors, such as pressurized water reactors (PWRs) and heavy water reactors (HWRs), fuel assemblies may be arranged in such a way that a blanket of mainly thorium fuel rods surrounds a more enriched seed element containing ^{235}U, which supplies neutrons to the subcritical blanket. As ^{233}U is produced in the blanket, it is burned there. This is the light water breeder reactor concept, which was successfully demonstrated in the United States of America in the 1970s. The CANDU HWR has also been tested extensively with thorium based fuels.

Currently, the thorium fuel cycle is again of interest to achieve improved proliferation resistance. In IAEA-TECDOC-1319 [III–1], an overview is given of thorium based nuclear fuel cycles (NFCs) in comparison with other fuel cycles.

The broad deployment of a commercial thorium nuclear fuel cycle depends on the progress of advanced reactor concepts, in particular an advanced high temperature gas cooled reactor (HTGR).

Thorium based fuels which could be used in the future include:

- Mixed plutonium–thorium oxide or mononitride for burning weapons grade or civilian plutonium;
- Mixed Th–^{233}U oxide or nitride for ‘clean’ energy production;
- Mixed highly enriched uranium (HEU)–thorium oxide or nitride for burning HEU.

It has been shown that the nitride fuel system burns plutonium more efficiently than does oxide fuel. The nitride fuel system produces about 20% more ^{233}U than the oxide fuel system but also produces a higher quantity of minor actinides. The oxide fuel energy amplifier system can incinerate 40% of the loaded plutonium in one cycle, while the nitride fuel system can destroy only 35%. However, given the necessity of using ^{15}N in the case of nitride fuels, the mixed oxide fuel system is preferred, especially for burning plutonium. The role of thorium based fuel in accelerator driven systems (ADSs) and in fast critical systems is more or less the same and there is no large difference in plutonium consumption.

The following concepts may have a chance to be realized within the next 25–50 years:

- Thermal reactors with fuel based on plutonium oxide (PuO_2), thorium oxide (ThO_2) and/or uranium oxide (UOX) (UO_2) particles arranged in fuel rods.
- HTGRs of two kinds: pebble bed and with prismatic fuel elements.
- Advanced heavy water reactor (AHWR): The main part of the core is subcritical with Th/^{233}U oxide, mixed so that the system is self-sustaining in ^{233}U. A few seed regions with conventional mixed oxide (MOX) fuel will drive the reaction and give a negative void coefficient overall.
- As an alternative to the MOX (U, Pu) fuels currently used in conventional light water reactors (LWRs), Th–Pu fuel may be used, with plutonium being consumed and fissile ^{233}U bred. The remaining ^{233}U after separation could be used in a Th–U fuel cycle.
- High conversion boiling water reactors, as under consideration within the GEN-IV advanced reactor concepts, using previous research on high conversion LWRs in Europe, Japan and the USA over the past several years [III–2], including advanced fuel designs such as metal matrix ‘dispersion’ fuel.
- An advanced HTGR (e.g. the French very high temperature reactor (VHTR)), which, according to the French Alternative Energies and Atomic Energy Commission (CEA), is planned for the production of heat and hydrogen. This reactor would operate with either TRISO UO_2–SiC fuel or (later) with ZrC coated particle fuel.
- Fast breeder reactors may use plutonium based fuel to breed ^{233}U from thorium, and then advanced nuclear power systems will use the ^{233}U.
- Pressurized heavy water reactors (PHWRs) with heterogeneous configurations of UO_2 and ThO_2 in separate elements in a bundle, or in separate fuel channels; alternatively, homogeneous (Pu–Th)O_2 bundles. In the longer term, ^{233}U/thorium would be recycled (with the self-sufficient equilibrium thorium (SSET) self-sufficient in fissile requirements).
- The molten salt fast reactor (MSFR), a concept that appeared in the twenty-first century, allows breeding from thorium and burning of plutonium and minor actinides. Two versions are foreseen: one for burning plutonium

and minor actinides (Russian MOSART) and a breeder version with ^{232}Th as the fertile isotope. This type of fast reactor is supposed to present a high degree of safety and a strong reduction of actinide waste.

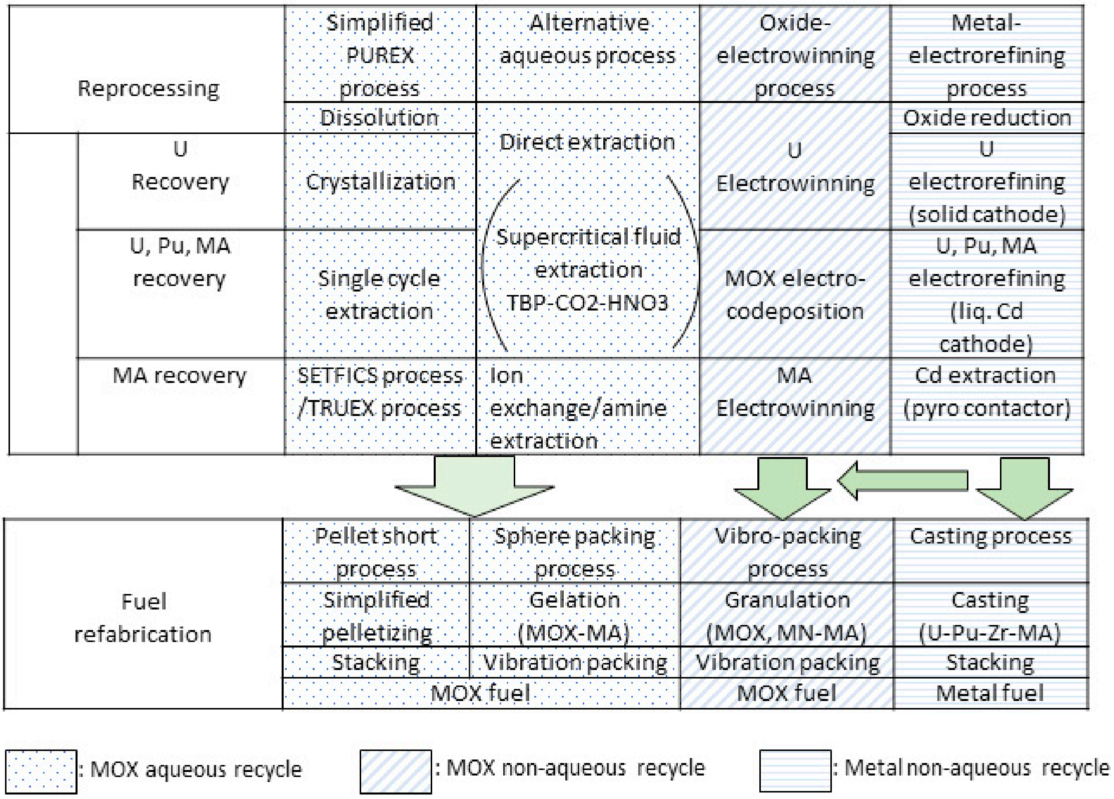
III–1. NUCLEAR FUELS FOR INNOVATIVE REACTORS

Some of the innovative reactors will use advanced fuels or have the ability to use them. Changes in fuel design may affect the generation of radioactive waste from fuel production as well as radioactive waste from spent fuel reprocessing. The need for development efforts in fuel design applies most obviously to lead cooled, molten salt cooled and thorium adaptable reactors, for which there is only limited experimental experience in fuel design and virtually none in the development of a commercial fuel cycle.

MOX fuel

In the past decade, a considerable amount of experience has been collected with the design, fabrication and performance of MOX fuel in Europe, particularly in France. Mixed uranium–plutonium oxides, containing up to 30% PuO_2 and UO_2 with HEU (≥85% ^{235}U), have been successfully used as driver fuels in most of the prototype fast reactors in the world. Industrial scale experience with fabrication, irradiation, reprocessing and re-fabrication of mixed oxide fuels has been established for thermal reactors. However, the use of mixed oxide as a driver fuel in commercial fast reactors is vulnerable mainly because of its low breeding ratio and long doubling time (>25 years). The low thermal conductivity of oxide fuel is also a disadvantage.

Advanced MOX fuel fabrication technologies (Fig. III–1), such as simplified pellet, sphere/vibro-packing and metal casting processes, are under development in Japan [III–3]. In the Russian Federation, two principal technologies are being developed to process the plutonium (mainly low burnup) into mixed uranium–plutonium fuel: pelletizing of U–Pu powder and vibro-compacting, which directly uses the acid solution from recycling and which could have economic merit compared with the classic pelletizing route.



Reprocessing	Simplified PUREX process	Alternative aqueous process	Oxide-electrowinning process	Metal-electrorefining process
	Dissolution	Direct extraction	U Electrowinning	Oxide reduction
U Recovery	Crystallization	Supercritical fluid extraction TBP-CO2-HNO3		U electrorefining (solid cathode)
U, Pu, MA recovery	Single cycle extraction		MOX electro-codeposition	U, Pu, MA electrorefining (liq. Cd cathode)
MA recovery	SETFICS process /TRUEX process	Ion exchange/amine extraction	MA Electrowinning	Cd extraction (pyro contactor)

Fuel refabrication	Pellet short process	Sphere packing process	Vibro-packing process	Casting process
	Simplified pelletizing	Gelation (MOX-MA)	Granulation (MOX, MN-MA)	Casting (U-Pu-Zr-MA)
	Stacking	Vibration packing	Vibration packing	Stacking
	MOX fuel		MOX fuel	Metal fuel

░: MOX aqueous recycle

▨: MOX non-aqueous recycle

▤: Metal non-aqueous recycle

FIG. III–1. *Innovative fuel fabrication technologies for fast breeder reactors in Japan. MA — minor actinide; MN — mononitride; MOX — mixed oxide.*

Carbide and nitride fuel

Mixed uranium–plutonium monocarbide (MC) and mononitride (MN) were identified as advanced fast reactor fuels nearly three decades ago on the basis of their high heavy metal density, high breeding ratio (and, in turn, short doubling time), high thermal conductivity and excellent chemical compatibility with sodium coolant. Monocarbides and mononitrides belong to the same family on the basis of their crystal structure and similar physical and chemical properties.

The monocarbides and mononitrides of uranium and plutonium have complete solid solubility. The international experience with carbide and nitride fuels has been very well documented in the proceedings of several international conferences and IAEA meetings [III–4 to III–11]. However, compared with mixed oxide fuel, the experience with monocarbide and mononitride fuels is very limited.

The major problem with mononitride fuel is the formation of radioactive ^{14}C by (n,p) reaction with ^{14}N and the high parasitic absorption of fast neutrons by ^{14}N. The problem with ^{14}C could be avoided by using ^{15}N. However, the process of ^{15}N enrichment is expensive. An alternative way to resolve the ^{14}C problem is to isolate it during reprocessing, oxidize it into CO_2 and finally convert it to $CaCO_3$ and bury it in a deep geological disposal facility as high active solid waste.

The different techniques of synthesis and consolidation of monocarbides and mononitrides are similar because these non-oxide actinide compounds are iso-structural, completely solid solubles and have very similar physical, chemical and thermodynamic properties. The main reasons that UC, PuC, (U,Pu)C, UN, PuN and (U,Pu) N are difficult and expensive to fabricate are: First, the greater number of process steps compared to those for oxide fuel. Second, these actinide compounds are highly susceptible to oxidation and hydrolysis and are pyrophoric in powder form. The entire fabrication is, therefore, required to be carried out inside leaktight gloveboxes maintained in an inert cover gas (N_2, Ar, He, etc.) atmosphere containing minimal amounts of oxygen and nitrogen (<20 ppm each). Third, stringent control of the carbon contents is needed during the different stages of fabrication in order to avoid the formation of the unwanted metallic phase and to keep higher carbides (M_2C_3 and MC_2) within acceptable limits. Higher nitrides (M_2N_3 and MN_2) dissociate to mononitrides at elevated temperatures (≥1400°C) in an inert atmosphere and pose no problem.

The two main steps for fabrication of UC, UN, PuC, PuN, MC and MN fuels are as follows [III–4]:

(1) Preparation of buttons, powders, clinkers or sol-gel microspheres of the monocarbide or mononitride, starting either from the oxide or from the metal;
(2) Consolidation of monocarbide or mononitride powders, granules or microspheres in the form of fuel pellets, followed by loading of the fuel pellet stack in a cladding tube and encapsulation or vibro-packing of granules or microspheres in a fuel cladding tube, followed by encapsulation.

Despite the fact that less research has been done into mononitride fuel compared with monocarbide fuel, mononitride has better prospects than monocarbide as a fuel for future fast reactors with a closed fuel cycle. This is because of the simpler fabrication technology of mononitride compared with carbide fuel, its superior thermo-physical properties and its suitability for reprocessing by the plutonium uranium redox extraction (PUREX) process and pyrometallurgical processes.

Metallic fuel

The first fuel used in fast reactors was a metal fuel because of its high thermal conductivity, high fissile density and the availability of experience with metallurgy [III–4]. These benefits were demonstrated at the experimental breeder reactor (EBR-II), where tests were conducted at full power with complete or partial loss of flow. Also, the metallic fuel lends itself to straightforward reprocessing by means of relatively simple and inexpensive electro-refining. However, metallic fuel was not chosen for mainline development in the 1960s because of its high burnup potential and because the cladding ability to operate at very high temperatures had not been demonstrated.

Programmes in the USA and partially in the Russian Federation were addressed to improve fuel and cladding materials and fuel rod design. Doping elements were selected to increase the solidus temperature of uranium or uranium–plutonium fuel and the eutectic temperature with stainless steel cladding materials. The best results were obtained for the U–Pu–Zr alloy. Improvements in stainless steel cladding materials were directed to reduce the swelling rate and, at the same time, to ensure sufficient irradiation creep rate to avoid fuel cladding mechanical interaction.

Solid fuel for transmutation of actinides

Burning of fissile actinides in nuclear reactors requires inactive materials which act as diluents in the austenitic steel or Zr based clad fuel pins in order to soften the high fission density and consequently, the high fuel temperature ('cold' fuel concept). At present, many variants of diluents are under consideration: homogeneous and heterogeneous diluents forming a solid solution with fissile material and a two phase microstructure consisting of ceramic and metallic diluents, respectively [III–12 to III–18]. The selection of diluents is based on particular physical and chemical properties which determine the preparation, handling, thermal and irradiation stability, interaction with coolant and the dissolution behaviour in acids during reprocessing.

Both PuN and PuO_2 are considered in Refs [III–12 to III–18] as ceramic diluents (CERCER), taking into account their physical and chemical behaviour. Also, AlN and ZrN are recommended as heterogeneous and homogeneous diluents, respectively, for PuN fuel. CERMET (ceramic–metal) of the PuO_2–M type (where M might be Al or Zr) and the PuN–M type (where M might be V, Cr, Mo or W) is now also being considered as a potential solution for plutonium utilization in LWRs and fast reactors, respectively.

Thorium solid fuel

At present, no commercial thorium fuel fabrication plants exist. However, the fabrication technology for the thorium based fuels — especially the oxides — is likely to be rather similar to that for the other oxide or mixed oxide fuels. For example, ThO_2 pellets for use as a blanket in fast breeder thorium reactors (FBTR) in India are fabricated using the conventional powder metallurgy technique of cold compaction followed by high temperature sintering, as is done for the use of ThO_2 bundles for flux flattening in PHWRs. Since ThO_2 has a very high melting temperature of ~3400°C, the sintering has to be done at temperatures above 1800°C to achieve a high >96% theoretical density. Additions of dopants such as MgO and Nb_2O_5 have been used to significantly reduce the sintering temperatures.

Most promising for the future use of thorium fuel is the 'coated particle concept' developed in Germany and in the USA for high temperature reactor (HTR) fuel. The coated particles used are spherical fuel kernels (oxide or carbide of uranium, thorium or plutonium) surrounded by layers of a coating material such as pyrolitic carbon or silicon carbide. Powder agglomeration processes or wet chemical processes (sol-gel) for the gelation of droplets from a solution containing thorium and uranium could be used to produce the kernels. A process based on 'sol-gel microsphere pelletization' has also been developed in Germany for fabrication of high density oxide pellets for HTRs. This is done at relatively low compaction pressure and low sintering temperature, avoiding dust generation. A thorium high temperature reactor was built as a demonstration plant for the use of thorium fuel in coated particles, but it was operated only for a short time.

Significant experience with coated particle thorium fuel was accumulated in the USA in the Fort St. Vrain HTGR; 2448 hexagonal graphite fuel elements using 26 000 kg of fissile and fertile material in TRISO coated fuel particles were produced. (This included almost 25 000 kg of thorium. It was irradiated at temperatures greater than 1300°C to a maximum burnup in the fissile particles of 16% fissions of initial metal atoms totalling approximately 170 000 MW·d/t HM.)

Today, India is most interested in using thorium fuel and has made thorium fuel fabrication and in-pile use one of its major developmental targets.

III–2. REPROCESSING OF SPENT NUCLEAR FUEL

The radiotoxicity of spent fuel decreases as the result of radioactive decay. Long term radiotoxicity is due primarily to actinides and a small number of fission products with very long half-lives. Transmutation of these long lived nuclides into shorter lived nuclides would reduce the long term radiotoxicity of the material. However, such transmutation would require processing the spent fuel, which would make the toxic elements more mobile. In addition, the quantity of actinides on or near the surface may be increased for a long time as they are processed, stored and cycled to reactors or ADSs. Passive safety of storage systems could be affected by the different chemical and physical nature of the materials containing the actinides. Finally, additional occupational exposure may occur and shielded or remote facilities may be necessary [III–19]. This increases the complexity of the process.

The innovative reactors cannot be separated from spent fuel cycle reprocessing technology and waste management requirements. They are all interrelated. The recycling options are derived from the different strategies of the nuclear fuel cycle and the future of waste management.

The fuel reprocessing technologies under development in support of innovative reactors also have potential for nearer term application to optimize the capacity and performance of a geological repository. Recycling, fuel treatment, conditioning and transmutation technologies have the potential to dramatically reduce the quantity and radiotoxicity of waste requiring geological disposal. These technologies are not alternatives to geological disposal but help reduce the cost and optimize the use of a geological repository. Finally, the minimization of spent fuel and waste is a big challenge to future NFC technologies. Improvements in reprocessing need to be incorporated into the waste management system. Waste strategies to minimize the hazard from radioactive waste need to be implemented in all parts of the nuclear fuel cycle.

Originally, both aqueous and pyro processing were applied, primarily for military purposes (i.e. the PUREX process to extract sufficiently pure plutonium and the electro-refining process to clean plutonium metal from ingrown americium). This latter process was extended to reprocess metallic spent fuel from the EBR-II reactor at the Argonne National Laboratory (ANL) West [III–12]. In contrast with conventional aqueous processing, pyro processing does not involve dissolving spent fuel in an acid solution. Rather, the fuel is chopped and suspended in baskets in a molten salt bath through which an electric current flows. Most of the spent fuel constituents, including uranium, transuranics (TRUs) and fission products, dissolve into the salt. Whereas most of the fission products remain in the salt, uranium and TRUs are removed from the salt through deposition on different cathodes.

Reprocessing of oxide fuel

Aqueous reprocessing

Aqueous reprocessing of fast reactor fuels based on oxide, carbide and nitrides has been achieved mainly through the PUREX process, which utilizes tri-n-butyl phosphate (TBP) as the extractant for separating uranium and plutonium (see Annex I). TBP has been the mainstay of the process and has been universally accepted as a nearly ideal extractant. However, a few features of TBP — such as its significant solubility in the aqueous phases (which may lead to safety related problems during evaporation), tendency to degrade with products not easily amenable for treatment, tendency for third phase formation during the extraction of plutonium and issues related to ultimate disposal of the extractant — have provided the impetus to develop alternative extractants to replace TBP. Since the 1980s, alternative extractants based on amides have been studied in many countries, especially France, as replacements for TBP. In addition to possessing extraction properties similar to those of TBPs, the amides have other advantages, such as low aqueous solubility and the possibility of uranium–plutonium partitioning, without the reduction of plutonium to a lower oxidation state. The amides contain only atoms of the elements C, H, N and O, and are therefore completely combustible. However, amides have yet to be deployed even in reprocessing plants for thermal reactor fuels, and considerable experience on an industrial scale is necessary before they are accepted as a replacement for TBP.

Alternatives to PUREX aqueous methods, such as COEX, DIAMEX-SANEX and GANEX, are under development in France [III–20, III–21]. COEX is aimed at the extraction of all plutonium and a part of uranium for the manufacture of MOX fuel. GANEX and UREX+ in the USA are aimed at the extraction of pure uranium and the mixture of plutonium and other actinides.

Aqueous processes are often mentioned for the treatment of advanced fuels, but innovative processes like dry processes or pyro processing are promising.

Pyro processing

Among pyro processing methods is a fluoride volatility method. The existence of volatile fluorides of uranium, neptunium and plutonium prompted the development of fluoride volatility processes for the treatment of spent fuel in the 1960s and 1970s. The basis of this method is the ability of uranium and plutonium to form volatile UF_6 and PuF_6, while the main fission products remain in the non-volatile state.

During the 1980s, the Nuclear Research Institute of Czechoslovakia operated a small scale fluoride volatility processing line for the treatment of oxide fuel discharged from the Russian BOR-60 fast reactor [III–22]. A flame fluorination method was used in the process, and reasonable recovery efficiencies were achieved. The same institute, now in the Czech Republic, is studying the use of the fluoride volatility process for treatment of LWR spent fuel, in order to provide feed materials to an accelerator driven molten salt reactor (MSR) for consumption of transuranic elements (TUEs).

Fluorination of irradiated UOX was also studied on the spent fuel from the fast breeder BOR-60 reactor at RIAR, Russian Federation [III–23, III–24].

Pyroelectrochemical process in the chloride melts

An innovative process for oxide spent nuclear fuel (SNF) reprocessing has been developed in the Russian Federation [III–25]. This process includes three stages:

(1) Transfer of SNF components to the molten salt KCl, LiCl, CsCl at 600–700°C (dissolution);
(2) Removal of fissile materials (UO_2, PuO_2) with the deposition on the hard cathode;
(3) Procedures for concentration of fission products and recycling of reagents.

The UO_2–PuO_2 deposit on the hard cathode (pyrographite) is cooled, crushed, washed, dried and classified.

Reprocessing of mixed (U, Pu)O_2 fuel by recrystallization in molten molybdates

The method of MOX fuel reprocessing consists of its dissolution (after de-cladding) in the MoO_3–Na_2MoO_4 melt followed by precipitation of the homogeneous solid solutions (U, Pu)O_2 by cooling. The fission products remain in the melt and the crystals are separated. The purification from fission products as large as two to three orders of magnitude has been achieved.

The crystals of (U, Pu)O_2 are washed by Na_2MoO_4 melt, then (after cooling) by water, dried and directed into the remote manufacture of fuel elements. The melt is used many times. As a result of many cycles, the melt accumulates large amounts of non-volatile fission products. From this melt, the oxides of fission products are precipitated by gradual adding of alkaline metal oxides or carbonates, or by direct recrystallization [III–26]. These oxides represent mainly high level waste (HLW).

Transfer of U–Pu from molten salts to molten metals

Calcinated products from the PUREX process after fluorination are dissolved in CaF_2–MgF_2 and the actinides contained in molten salts are transferred into the metallic melt (Zn, Cu–Al, Cu–Al–Zn) in the presence of a reducing agent (Mg). The solvent is then removed by distillation. It also appears possible to chlorinate the calcinated product and then recover the minor actinides by electrochemical means.

A *pyrochemical* reprocessing method for spent LWR oxide fuel (PYRO-A) has been studied at the ANL with the goal of separating TRU and fission products. After de-cladding and crushing, the fuel is reduced to a metallic state by electrolysis in a LiCl bath containing 1 wt% Li_2O [III–26]. Uranium is extracted by an electro-refining process. The metallic uranium is deposited on a solid steel cathode, and the uranium is recovered by melting at reduced pressure to volatilize off any adhering LiCl. The TUE and fission products are anodically dissolved and remain in the LiCl electrolyte salt. The TRUs are recovered on a cathode and form ingots for the preparation of transmutation targets.

Reprocessing of carbide and nitride fuels

A PUREX process has been tested with regard to reprocessing monocarbide and mononitride fuels in India, the Russian Federation [III–27] and Europe [III–28]. From these preliminary experiments, it appears that the PUREX process developed for MO_2 fuels can be applied without any change to MN fuels.

A PUREX process can also be applied to the advanced fuels (carbides, carbo-nitrides, nitrides) after their conversion to oxide form. Conversion can be done using a wet chemical process, but this involves the production of complex organic molecules which interfere with the extraction process. A gaseous oxidation process does not have this disadvantage [III–29, III–30]. For easy reprocessing, the oxidized fuel must be exclusively close to stoichiometric (U, Pu)O_2 containing 25% Pu. Samples of advanced fuels have been oxidized in gaseous environments to determine the most appropriate means of achieving the required degree of oxidation.

Alternatively, the reprocessing of mononitride fuel based on pyrometallurgical techniques, used before for metallic fuel, has been studied in the Russian Federation [III–31] and Japan [III–32 to III–35]. This method uses the electrochemical dissolution of mononitride fuel to recover metal uranium and plutonium as precipitates on a cathode. This method has been demonstrated for reprocessing unirradiated UN. The feasibility of reprocessing irradiated material has yet to be demonstrated.

The reprocessing of the nitride spent fuel from fast reactors should include dismantling the fuel assemblies: separating the fuel, cladding and lead (if lead is used as a coolant). The efficient separation of fission products is realized by anode dissolution of fuel pellets and cathode deposition of uranium and plutonium metals at 500–550°C. The mixture of melted KCl, LiCl, UCl_3 and $PuCl_3$ salt is used as an electrolyte.

The uranium–plutonium mixture is sent for melting and uranium content correction. The corrected uranium–plutonium alloy will be used to produce nitrides. The electrolyte may be used repeatedly. During the anode dissolution of the fuel, some insoluble impurities (noble metals, molybdenum, zirconium and technetium) remain as slurry; others (rare earths, alkalines and alkaline earths) will be dissolved and transferred into the electrolyte as chlorides. During electrolysis, gaseous products such as krypton, xenon, tritium and nitrogen will be emanated as well as iodine and aerosols. After the fission products accumulate in the electrolyte, these products will be recovered, partitioned (if necessary) and sent to the reactor for transmutation or to a long term storage facility after certain conditioning. The purified electrolyte will be reused [III–36]. Metals are used to separate nitride fuel from its stainless steel cladding and to melt and dissolve the classing at a low temperature.

Reprocessing of metal fuel

Metal fuel may be reprocessed using the pyrometallurgy (electro-refining) method which was developed for metal fuel in the USA and used for reprocessing EBR-II fuel assemblies. The key step in the pyrometallurgic reprocessing of metal fuel is electro-refining [III–22]. A schematic view of the electro-refining process is given in Fig. III–2.

In the electro-refining method, metallic fuel pins are chopped and the fuel pin segments placed in a stainless steel mesh basket that becomes the anode of an electro-refining cell using a LiCl–KCl electrolyte. Uranium is electro-transported from the salt to the steel cathode. The TUE will not deposit at the steel cathode as long as the transuranic–uranium ratio in the salt is less than 100 or so. Recovery of the TUE requires the use of a different cathode, one in which the TRUs deposit as inter-metallic compounds with cadmium in a crucible containing liquid cadmium that is suspended in the electrolyte salt. Deposition of the TRUs is accompanied by a certain amount

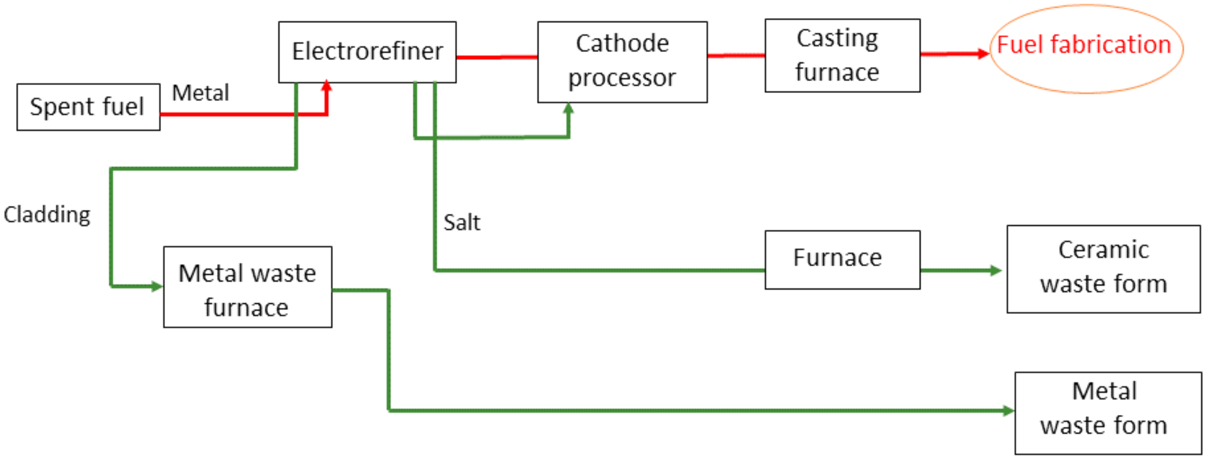


FIG. III–2. The pyro processing flowsheet [III–22].

of uranium, depending upon its concentration in the salt. A mixture of 70% transuranics, 25% uranium and 5% lanthanide fission products is typical.

A modified version of the electro-refining process is used in the treatment of EBR-II fuel and blanket elements for disposal. Because this treatment is for the purpose of waste management and eventual disposal of HLW, only uranium is recovered. The TUEs are left in the electrolyte salt, which will be periodically removed to produce a composite glass–ceramic waste form containing the TRUs and the active metal fission products. The noble metal fission products that remain in the anode basket are combined with the cladding hulls and melted together to form a metallic waste form for disposal.

Two electro-refiners are used for the treatment of EBR-II fuel (~1.5 t) and blanket (~29 t) at the ANL. The highly enriched driver fuel is processed in an electro-refiner with a batch size of 20 kg, whereas the depleted uranium blankets are processed in an advanced low resistance electro-refiner with a 350 kg capacity. Both systems are being operated successfully, and the process of spent fuel treatment will continue for several more years. Even though there is very little fission product content in the blanket fuel, there is a significant quantity of plutonium present, and it may be necessary to remove the plutonium from the electro-refiner electrolyte salt before the fission product content reaches a level that would be appropriate for salt removal. About 2.4 t of EBR-II fuel was processed by this method over a five year demonstration period. The cathode product contains uranium, plutonium and minor actinides along with residual fission products. However, fission products can be adequately separated, and this process can produce satisfactory reprocessed fuels allowing further nuclear use.

This reprocessing technology has several benefits. First, diversion of the fuel is impossible since the material is highly radioactive; likewise, the process makes proliferation of nuclear weapons unfeasible because the cathode product remains alloyed as well as radioactive. Second, the process involves batch operations and thus is easily scaled to meet local or increased reactor requirements. Furthermore, comparative cost analysis has shown the process to be very competitive over other reprocessing options. Finally, this process allows essentially all the actinides to remain in the fuel cycle, to be fabricated back into the fuel and fissioned. As a result, the HLW that emerges from this process will decay to background radiotoxicity levels in only hundreds of years, rather than a million years.

The Central Research Institute of the Electric Power Industry (CRIEPI) in Japan has followed a similar path and has performed work on actinide/lanthanide extraction in molten chloride and liquid metal (Cd, Bi) media by electrodeposition and liquid–liquid extraction [III–22]. In Japan, a pilot plant with the throughput of 50 t/a is being designed. The plant for the pyrochemical reprocessing on the industrial level is scheduled by 2025 [III–37, III–38].

The work was conducted in Japan with uranium and at the European Union’s Institute for Transuranium Elements in Karlsruhe, Germany, with plutonium. Scientists at the CEA Marcoule laboratory in France are conducting studies with liquid metal cathodes and with reductive extraction processes.

Management of coated particle fuel

Future gas cooled reactors will almost certainly utilize coated particle fuel. This fuel consists of fuel microspheres (UOX, MOX, U–Th oxides, etc.), 50 to 500 μm in diameter, with successive coatings of porous carbon, pyrolytic graphite, silicon or zirconium carbide, and a final outer coating of pyrolytic graphite. The overall particle diameter is on the order of 800 μm to 1 mm.

Some unique problems exist for the storage, transportation and disposal of spent fuel from a prismatic VHTR design. The volumes of fuel to be disposed of are relatively large if the complete prismatic blocks containing the fuel are disposed of. If the fuel pellets are removed from the prismatic blocks for separate disposal, specialized techniques will be required to limit the release of radioactive materials (e.g. ^{14}C). Even if recycled (the number of fuel cycles for a prismatic block is limited to three or four by physical changes in the graphite), the graphite prismatic blocks will ultimately have to be disposed of. The power density and heavy metal loading for pebble bed fuel is typically lower than in the compacts and prismatic blocks, so the volumes are similar. The relatively high cost of reactor grade graphite (currently estimated to be in the range of $75 per kilogram) may eventually justify reprocessing the graphite.

VHTR spent fuel is ideally suited for both long term storage and permanent disposal in a repository. The properties of spent thorium fuel as a waste product have been studied. It was found that the high degree of chemical stability and the low solubility of thorium make irradiated thorium based fuels attractive as waste forms for direct geological disposal. The TRISO fuel particle coating system, which provides containment of fission products under reactor operating conditions, also provides an excellent barrier for containment of the radionuclides for storage and geological disposal of spent fuel. Experimental studies have shown that the corrosion rates of the TRISO coatings are very low under both dry and wet conditions. The coatings are ideal for a multiple barrier waste management system. The measured corrosion rates indicate that the TRISO coating system should maintain its integrity for a million years or more in a geological repository. If the fuel is to be reprocessed (and requirements on radiotoxicity reduction may mandate such reprocessing), these coating layers must be removed in order to provide reagent access to the fuel material. Both aqueous and non-aqueous processes are being developed, initially in concept only, for the treatment of these fuels.

Non-aqueous processes now being studied for application to the treatment of coated particle fuel include fluoride and chloride volatility processes, carbo-chlorination processes and direct electrochemical dissolution. Due to the unique structure of the particle fuel, the difficulties essentially involve gaining access to the uranium kernels coated by carbon and SiC layers and dispersed in a large volume of graphite. Starting from past experience in this field, the CEA has recently initiated a research programme to propose attractive solutions. The mechanical extraction of compacts from the spent fuel blocks appears to be a promising approach, as well as removing the graphite from the compacts by pulsed currents to free the particles. Subsequent removal of the carbon and silicon carbide layers by high temperature oxidation or by carbo-chlorination to access the kernels is being assessed [III–39].

In the case of pebble bed fuel feasibility, tests showed promise in fragmenting fuel spheres and coating fuel particles using welding techniques. These techniques should also be effective for the fuel compacts of the prismatic fuels. After the fuel particles are fragmented, the fragmentation product can be fed directly into classical aqueous reprocessing in which the fuel dissolves in nitric acid.

A problem common to all of these conceptual processes is the disposition of the large amount of carbon and silicon remaining from the processing of this fuel. Waste volumes can be very large, even though the fuel burnup capability might be quite high. France and the USA are presently collaborating on the evaluation of coated particle fuel treatment processes, and there may be other collaborative efforts in the future. The experience of physically separating the carbon from uranium–thorium kernels in thorium high temperature reactor (THTR) fuel spheres at Dounreay demonstrated a real problem with fine carbon particulate in the glovebox. This was unirradiated fuel and it is debatable how a highly active cell would cope with the fine carbon dust produced, either in the ventilation system or in maintaining containment during filter and manipulator maintenance. For actinide recycling, gelation appears to be a suitable process for fabricating the kernels.

Reprocessing of thorium based solid fuel

Most of the constraints associated with the thorium fuel cycle involve reprocessing the spent fuel and the subsequent handling of the separated ^{233}U and thorium products. Recovery of ^{233}U alone or both ^{233}U and thorium has been developed and demonstrated on a pilot scale in the past. But the Thorex process has not yet achieved the same maturity of the Purex process. More experimental validations are needed to make the Thorex process robust. The residue of isotopically altered plutonium in the spent fuel would add considerably to the complexities and necessitate modifications in the basic scheme employed in the Thorex process. The THOREX process for the recovery and recycling of ^{233}U and Th has several special requirements that need attention. In contrast to UO_2, crystalline ThO_2 and PuO_2 are difficult to dissolve in nitric acid without adding appropriate amounts of fluoride. But the use of fluoride is undesirable because of its corrosiveness and incompatibility with common reprocessing equipment.

When ^{239}Np is formed as an intermediate to ^{239}Pu, it has a half-life of only 2.3 days as opposed to the 27 day half-life of ^{233}Pa (the intermediate product of ^{233}U). This necessitates long cooling periods prior to reprocessing; this in turn will increase the out of pile inventory. The (n,2n) reactions encountered during the irradiation of thorium lead to the formation of long lived ^{231}Pa and relatively short lived ^{232}U (68.9 years) with its hard beta gamma emitting decay products. Thus, ^{233}U produced in the reactor is contaminated with ^{232}U and the level of contamination depends on the isotopic composition of the initial thorium fuel, the burnup and the neutron spectrum encountered in the reactor. The radioactive contamination from ^{232}U in the separated ^{233}U product and from ^{229}Th and ^{228}Th in the separated thorium product will have to be taken into consideration while handling these products. The ^{231}Pa is the main long lived actinide that needs to be assessed for its long term environmental impact during Thorex HLW treatment and disposal. The radiological hazard in handling the reprocessed ^{233}U arises mainly from the alpha and gamma activities associated with the ^{232}U isotope formed during irradiation, and its short lived decay products.

Dry processes that have been developed for uranium–plutonium based fuels have been thought of as a way of avoiding the problems inherent in aqueous reprocessing. The possibility of using a fluoride volatility process is ruled out because thorium fluoride is non-volatile. The other possible option is to bring into solution the thorium based fuel in a molten salt and subsequently use electrochemical processes to recover the actinides. However, the behaviour of thorium in the oxide electro-winning or molten salt electro-refining processes with respect to uranium and plutonium has yet to be established experimentally in order to arrive at a possible flowsheet.

Reprocessing of transmutation fuel

A variety of different non-fertile fuels are being considered for use in partitioning and transmutation systems [III–40]. Fuels currently under study include metallic (alloy of TRU with Zr), nitride CERCER (dispersion of TRU nitrides in an inert matrix such as ZrN), oxide CERCER (dispersion of TRU oxides in inert ZrO_2 or MgO), CERMET (dispersions of TRU oxides or nitrides in a metal matrix such as Zr) and carbide CERCER (dispersion of TRU carbide in SiC). The high content of transuranics in these fuels, together with the high zirconium content in many of them, tends to favour non-aqueous reprocessing methods.

The development of processes for treatment of these fuels is just beginning, and it is a fertile area for international collaboration. There are already some indications that direct molten salt electro-refining is technically feasible for the metallic alloy fuel and for the nitride CERCER fuel. A similar process for the carbide CERCER is problematic because the liberated carbon would foul the electrolyte. To reprocess the transmutation fuel from fast reactors, a new process called PYRO-B is under development [III–21]. Originally, this process was developed for metallic fuel and later was suggested for oxide fuel.

In the near term, the technologically mature aqueous processing methods constitute the main path forward, capitalizing on the success of the well established PUREX process, while dry processes are considered adjunct or backup processes. In the longer term, however, fuel cycle applications related to advanced reactor concepts (LMFR, GCFR, MSR, etc.) may favour the use of pyrochemical processes which do not have the drawbacks discussed above [III–23].

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Annex IV

EVOLUTION OF SPENT FUEL REPROCESSING IN FRANCE

The development of the future fuel cycle in France is guided by the following considerations:

- Minor actinides (americium, neptunium and curium) will be recycled exclusively in the innovative reactors or in accelerator driven systems (ADSs). In this context, the two minor actinide transmutation routes (i.e. homogeneous recycling in the core with the mixed oxide (MOX) driver fuel or heterogeneous recycling in blankets or dedicated fuels) are still open.
- To limit the industrial risk for deployment by around 2050, the processes and technologies of the next fuel cycle are only an evolution of the current La Hague and MELOX processes and technologies. The implementation of an improved plutonium uranium redox extraction (PUREX) process is therefore needed for the multi-recycling of uranium and plutonium. The processes dedicated to multi-recycling minor actinides will be gradually deployed after that. A scenario of deployment of these new recycling facilities is given in Fig. IV–1.
- To increase the resistance of the fuel cycle to proliferation risk, the new reprocessing facilities will be integrated and implemented so as to produce no pure plutonium at any part of the fuel cycle. All operations of reprocessing and fabrication are realized in the same facility: no transport by road of fissile materials, reception of spent nuclear fuel (SNF) at one end of the plant and supply of fresh fuel ready to be irradiated at the other end.

To ensure the durability of the fuel cycle with a gradual deployment of actinide recycling, and using evolutionary existing technologies, the PUREX process has been improved, from the dissolution of SNF to the manufacturing of the MOX driver fuel. The COEX$^{\text{TM}}$ process (improvement of the PUREX process) allows the

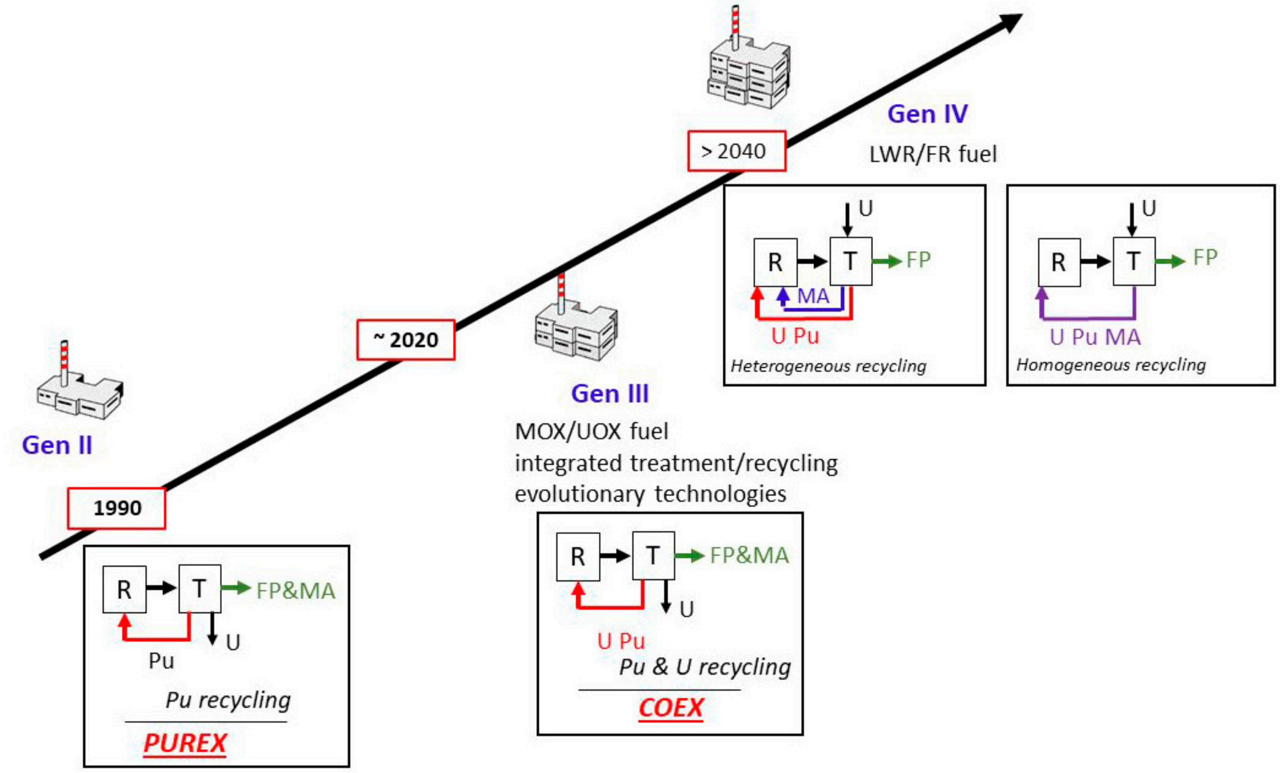

FIG. IV–1. Deployment scenario from existing to innovative fuel cycle facilities. FR — fast reactor; FP — fission product; MA — minor actinide; MOX — mixed oxide.

co-management of uranium and plutonium (and neptunium also, if needed) and the multi-recycling of these two elements (preservation of resources) in light water reactors (LWRs) and fast neutron reactors, while ensuring no pure plutonium is in the fuel cycle (increasing the first barrier against the proliferation risk).

The goal of recycling at least 99% of americium, neptunium and curium from SNF has quickly oriented research towards a separation of the long lived radionuclides (LLRNs) by aqueous processes from the raffinate of the PUREX process. This option needs the development of new specific extractant molecules that are different from the tri-n-butyl phosphate (TBP) used for the PUREX process. The advantage of this option is the implementation in the liquid–liquid contactors adapted for the nuclear environment. The process of separating the minor actinides from the raffinate of the PUREX, better known as the DIAMEX–SANEX process, was implemented in the liquid–liquid contactor technology used in La Hague (pulsed column, mixer settlers and centrifugal extractors) throughout the treatment of 10 kg of SNF in 2005. This technical feasibility demonstration was successfully followed by tests on other unique solutions of other minor actinide separation processes developed in France between 2006 and 2010 (i.e. TODGA SANEX, EXAM) based on the same approach to separating minor actinides from PUREX or COEX$^{\text{TM}}$ raffinate. Figure IV–2 shows the panel of available processes that are compatible with a mixed mode of actinide recycling (i.e. americium and curium together or americium alone) which can be added on-line to the PUREX and COEX$^{\text{TM}}$ processes.

The recycling mode for grouped actinides recommended by the Generation IV International Forum (GIF) encouraged France to develop the GANEX process in which uranium, plutonium, neptunium, americium and curium are separated together. A representation of the concept is given in Fig. IV–3. The most successful part of this concept against the risk of proliferation is that it replaces, rather than merely supplements, the previous concepts (i.e. PUREX, COEX$^{\text{TM}}$ DIAMEX, SANEX), which were developed to gradually introduce the multi-recycling of actinides compatible with the treatment of SNF coming from mixed power reactors (LWRs and fast reactors). Faced with the difficulty of having a selective extraction molecule for the grouped separation, mainly due to the presence of actinides at different oxidation degrees from III to VI, uranium is now separated at the beginning of the cycle by a specific extractant of An(VI) and the DIAMEX–SANEX process for the co-extraction of other actinides,

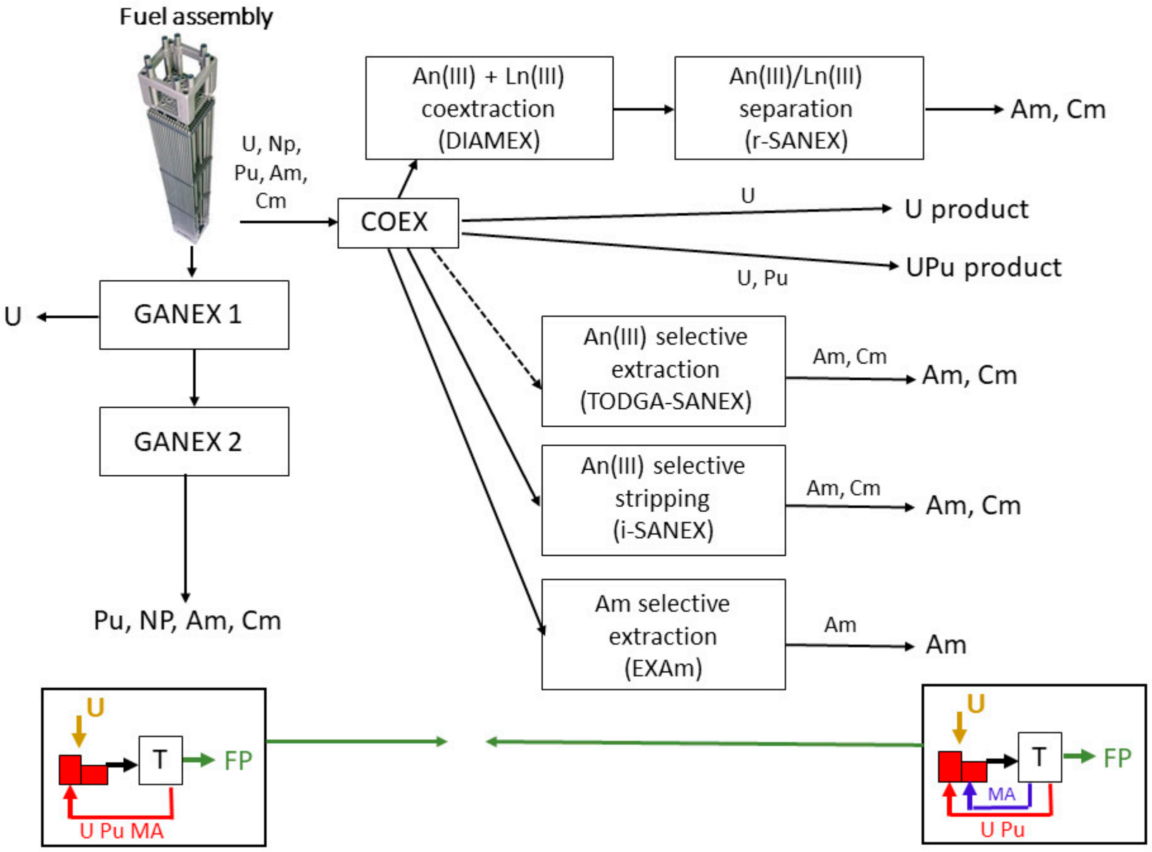

FIG. IV–2. French minor actinides separation processes. FP — fission product; MA — minor actinide.

such as plutonium, as well as minor actinides is managed. The PUREX process developed by the Argonne National Laboratory (ANL) (see Fig. IV–4) uses the same approach but not the same extraction system. Many countries are currently working on the integral separation of actinides, such as NUEX (new uranium extraction process) in England, NEXT (new extraction system for transuranic (TRU) recovery) in Japan, and REPA (recycling plutonium and minor actinides) in the Russian Federation.

Using the same approach to a gradual multi-recycling of actinides and the choice to implement evolutionary existing technologies, the oxalic conversion process of plutonium oxide and the manufacturing of MOX fuels by metallurgy powder is extended to the oxalic conversion of mixed actinides and their manufacture into oxide fuels (i.e. $UPuO_2$ for the driver fuel, $UAmNpCmO_2$ for the heterogeneous actinides recycling mode and $UPuAmNpCmO_2$ for the homogeneous route). These different processes are now under qualification.

In summary, the choice to use the evolution of existing technologies for the treatment and manufacturing of fuel for innovative reactors should lead to an equivalent efficiency in the separation and conversion of actinides and fuel fabrication operations, and produce two similar waste forms: glasses and compact metallic packages.

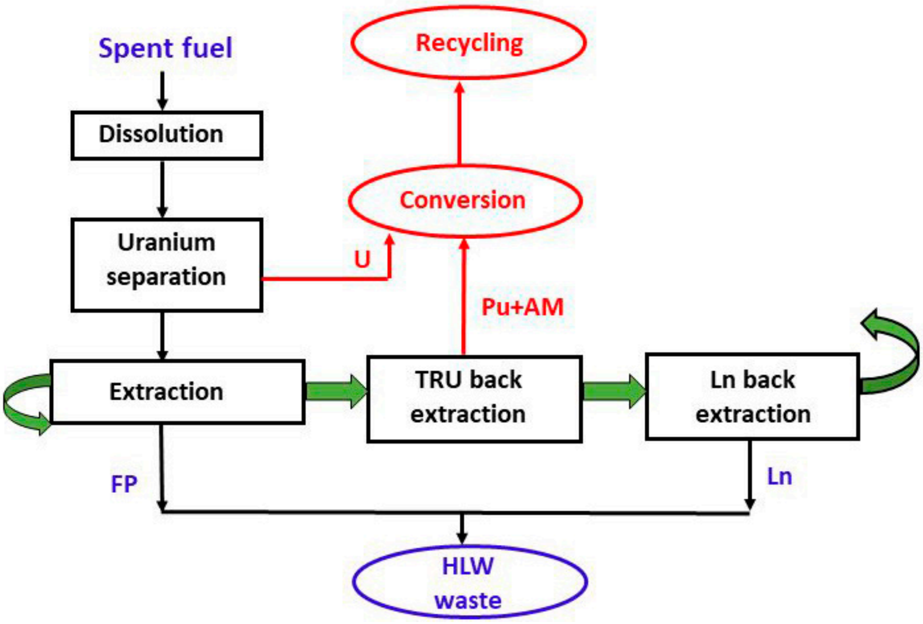

FIG. IV–3. GANEX concept for the homogeneous multi-recycling of actinides. AM — americium; FP — fission product; HLW — high level waste; TRU — transuranic.

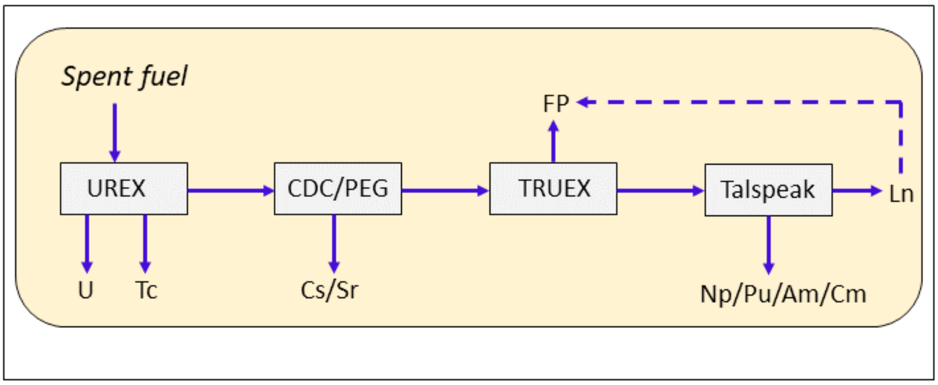

FIG. IV–4. UREX process for actinide recycling in a fast reactor. CDC — chlorinated cobalt dicarbollide; FP — fission product; PEG — polyethylene glycol.

ABBREVIATIONS

ADS	accelerator driven system
AHWR	advanced heavy water reactor
ANL	Argonne National Laboratory
GCFR	gas cooled fast reactor
GIF	Generation IV International Forum
HEU	highly enriched uranium
HLLW	high level liquid waste
HLW	high level waste
HTGR	high temperature gas cooled reactor
ILW	intermediate level waste
INPRO	International Project on Innovative Nuclear Reactors and Fuel Cycles
INS	Innovative Nuclear Energy System (INPRO)
L&ILW	low and intermediate level waste
LFR	lead cooled fast reactor
LMFR	liquid metal fast reactor
LWR	light water reactor
MOX	mixed (uranium–plutonium) oxide
MSFR	molten salt fast reactor
MSR	molten salt reactor
NFC	nuclear fuel cycle
NM	noble metal
NPP	nuclear power plant
PHWR	pressurized heavy water reactor
PUREX	plutonium uranium redox extraction
PWR	pressurized water reactor
R&D	research and development

SCWR	supercritical water cooled reactor`
SFR	sodium cooled fast reactor
SNF	spent nuclear fuel
TBP	tri-n-butyl phosphate
TRU	transuranic
TUE	transuranic element
UOX	uranium oxide
VHTR	very high temperature reactor

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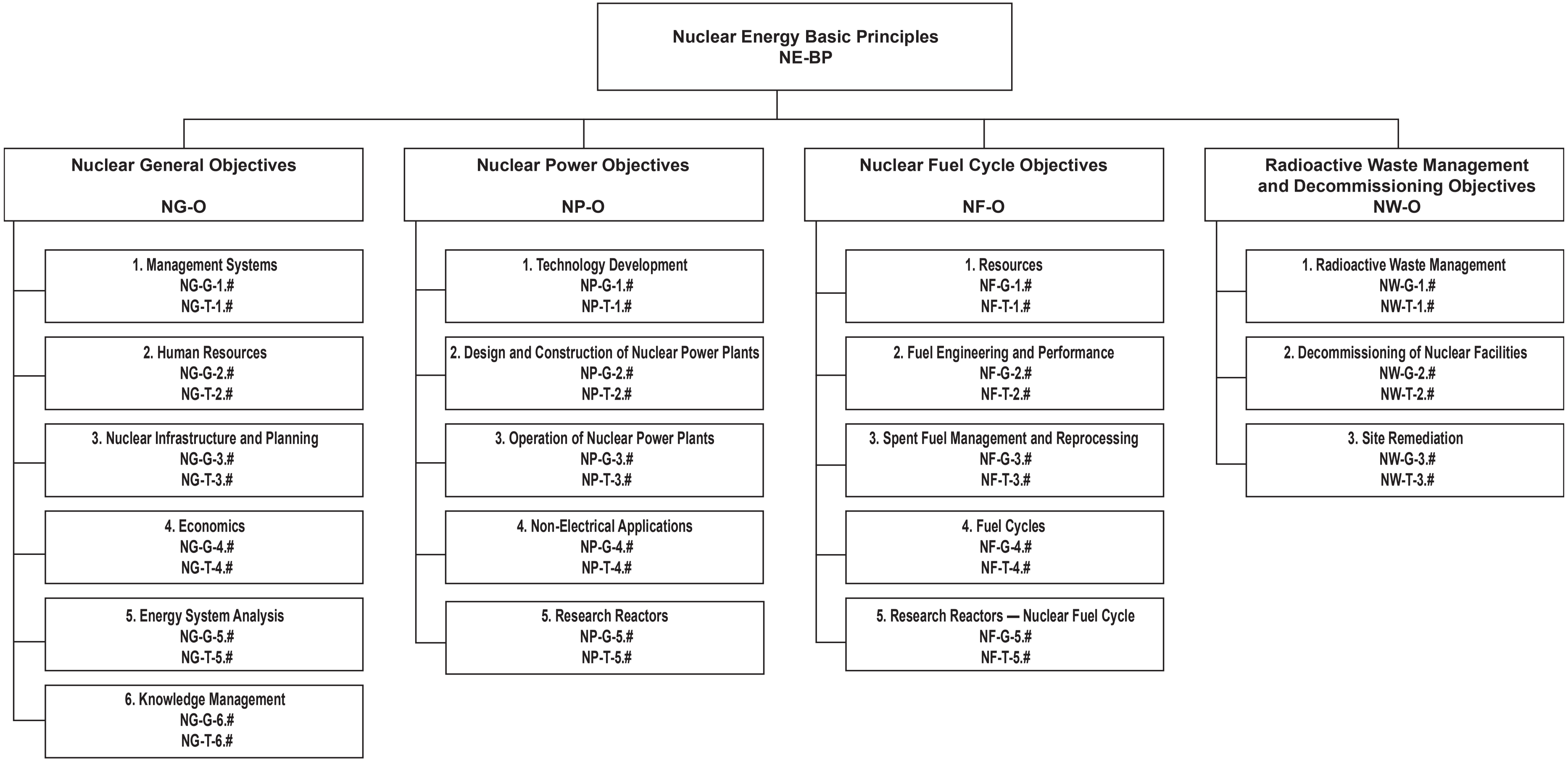

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